Radiation Effects on Plastic Scintillators for Current and Future HEP Experiments

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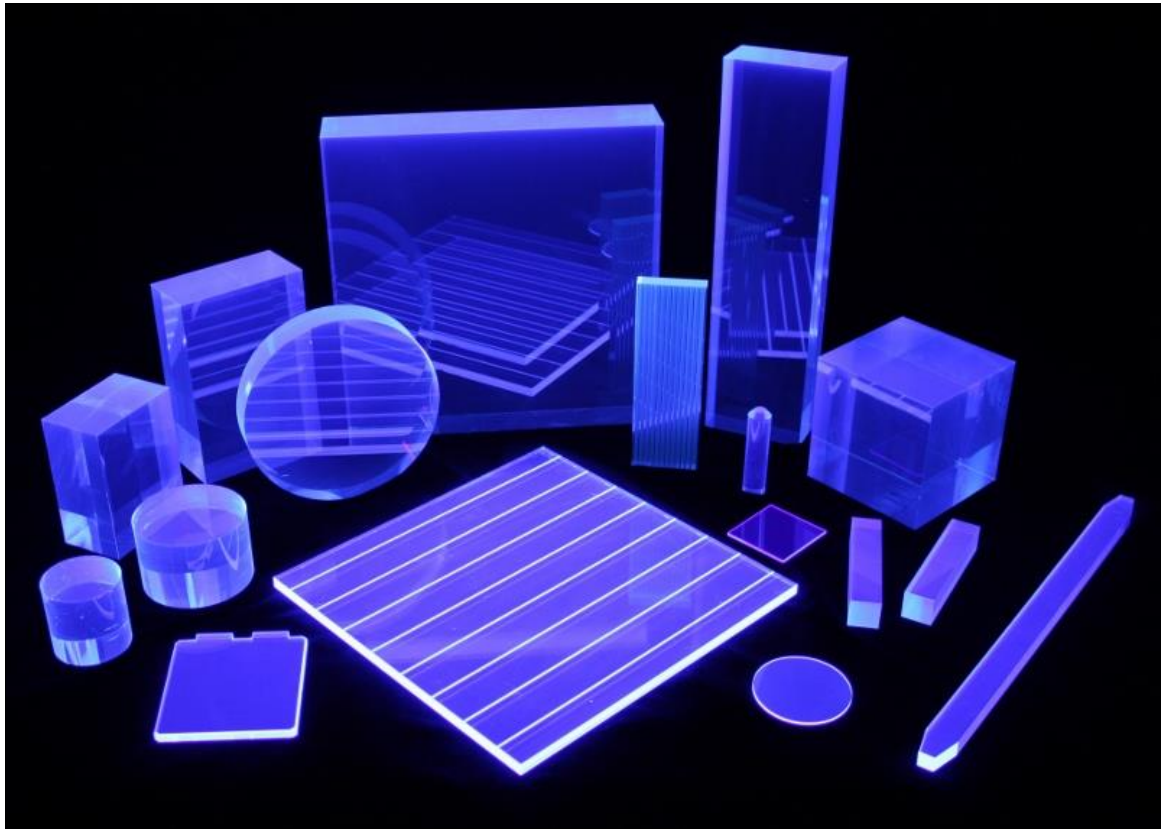
Research Techniques Seminar

Fermi National Accelerator Laboratory

November 20th, 2018

Plastic Scintillators in HEP

- Material of choice for hadron calorimeters of currently operating detectors
  - Commercially available in the large quantities needed for big detectors; plastic scintillators are cheap
  - They can be molded in any shape, provide design flexibility
  - They are fast: can provide info about energy in event in time for online selection
- Plastic degrades during irradiations
  - LHC detectors operate in unprecedented hostile conditions



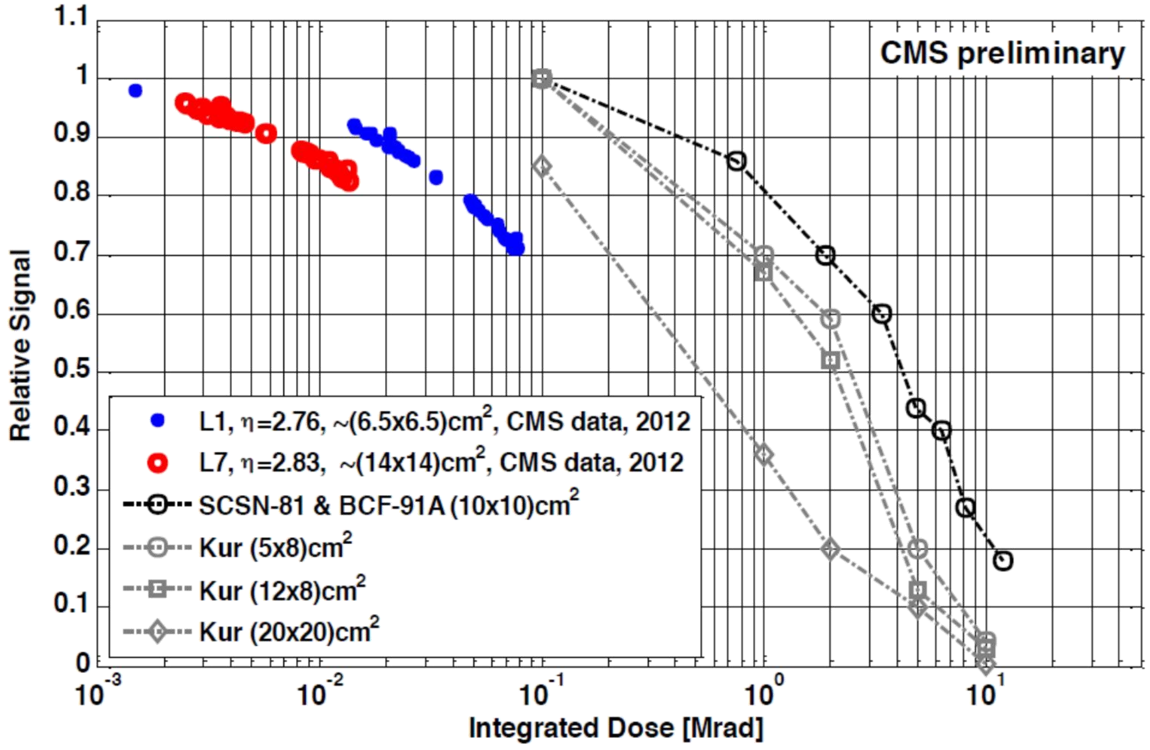
History of Scintillation Detectors

- 1903: Crookes builds first scintillation detector
  - A film of ZnS, scintillating when hit by an α particle; light detected by human operator (using microscope…)
- 1944: Curran and Baker introduce the PMT
  - Convenient replacement for naked eye; revives interest in scintillation detectors
- 1964: Birks “The Theory and Practice of Scintillation Counting”
- ~1990: SSC experiments raise the threshold for radiation tolerance
  - Many lessons taken (and some forgotten…) in design of LHC experiments

Ubi Crookes ibi lux

CMS HCAL Ageing

- The CMS Hadron calorimeter uses plastic scintillator as active material
  - It is know that radiation breaks the plastic and creates “color centers” which absorb scintillation light
- The crucial question: how long will it take the HCAL to become dark?
  - The lesson from 2012 data: shorter than it was originally thought
- R&D efforts aims at identifying a more radiation-tolerant material usable in HCAL upgrade and future detectors
  - Time scale: Long-Shutdown 3 upgrades (2024-2026)



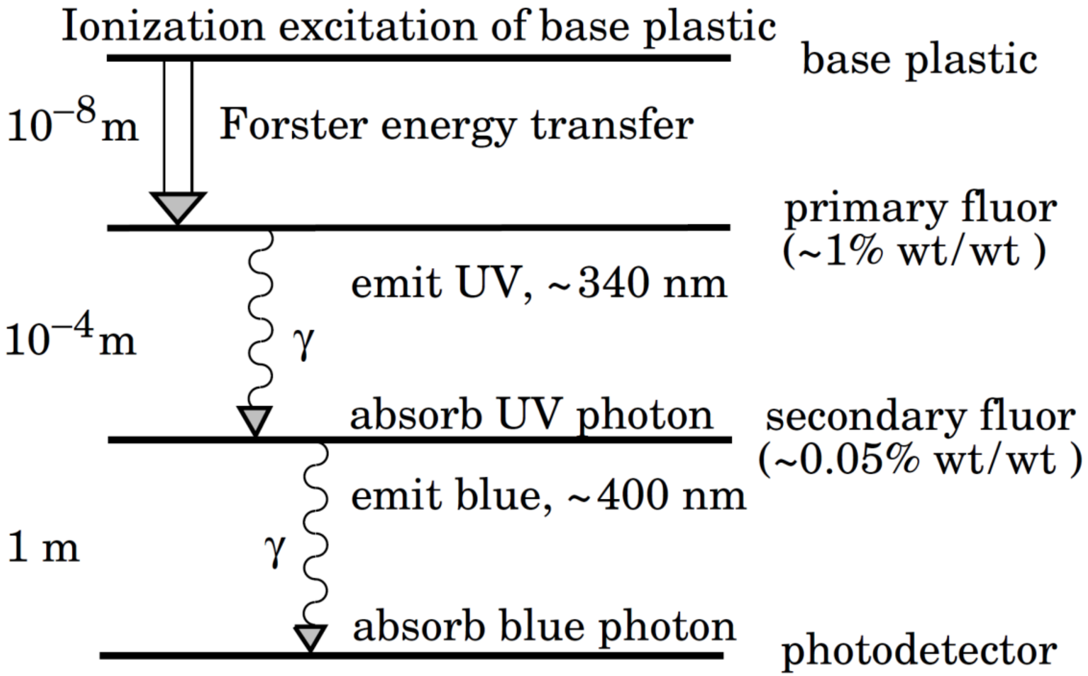
After an irradiation of 10krad, we see the light-yield reduction predicted for 1Mrad

Outline

- **How do plastic scintillators work?**
- **Measurements of radiation-induced damage, and their interpretation**
  - **Spectrophotometry, radioactive sources and cosmic rays**
  - **Irradiations with radioactive sources, LHC beamline**
- **Lessons learned**
  - **An attempt at putting together all the measurements**

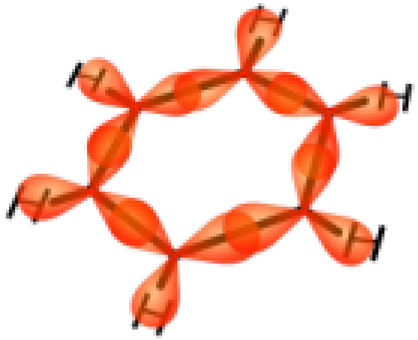
How does a Scintillator work?

- An organic scintillator is typically composed of three parts
  - A polymer base
    - Typically PVT, polystyrene, or silicon-based materials
  - A primary dopant (~1%)
  - A secondary dopant (~0.05%)
- Particles excite the base, the excitation of the base can migrate to the primary dopant, producing detectable light
  - In crystals, excitons transfer the energy; in liquids, solvent-solvent interactions and collisions
- The secondary dopant shifts the light to longer wavelengths, to make it more easily detected
  - Maximize the overlap with the wavelength range at which photodetectors are most efficient

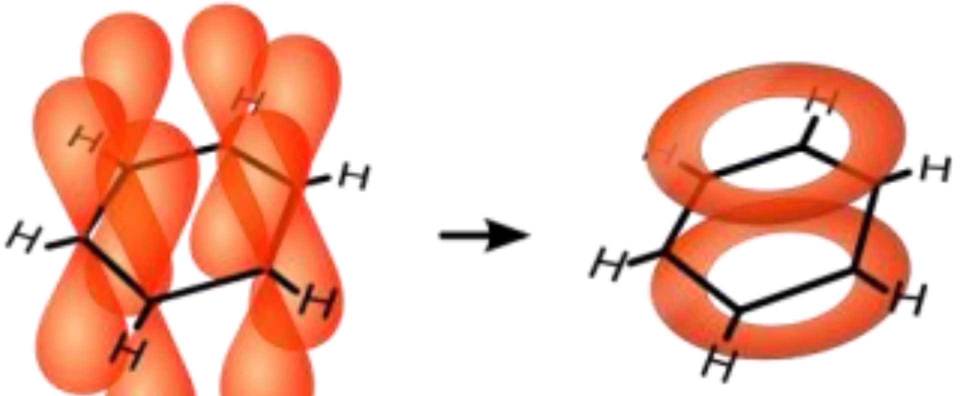


Chemistry Refresher

- Most common scintillator bases are PVT and PS, all carbon-based
  - The parts of interest are the C_6H_6 aromatic cycles
- Carbon atom has four external electrons, all participating in bond
  - One of $2s^2$ electrons promoted to 2p level
- The trigonal hybridization of sp^3 orbitals is luminescent
  - One p orbital untouched (π electrons), the other sp^2 orbitals mix into shared orbitals, at 120 degrees (σ electrons)
- At leading order, the light yield of the base is proportional to the ratio of π to σ electrons
  - More complex monomers enter the picture at NLO
  - Maximal LY reached by anthracene $C_{14}H_{10}$



Sigma Bonds
sp^2 Hybridized orbitals

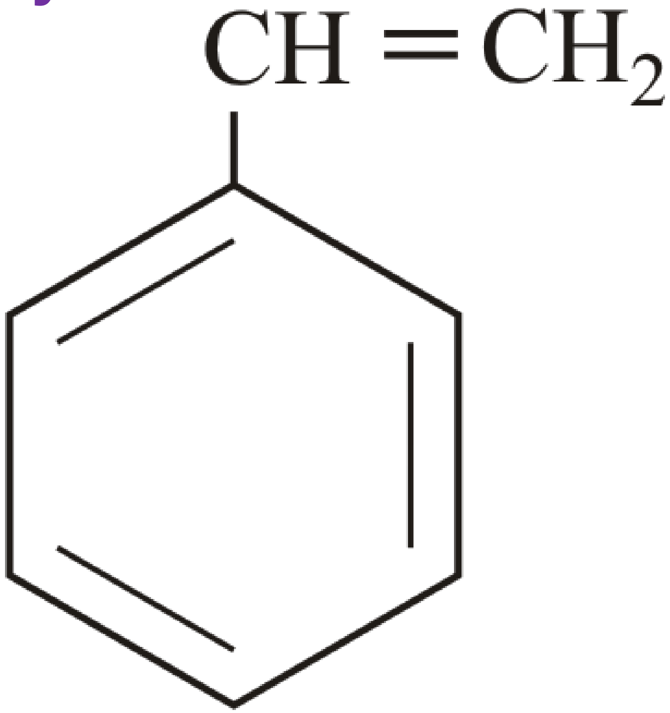


6 p_z orbitals

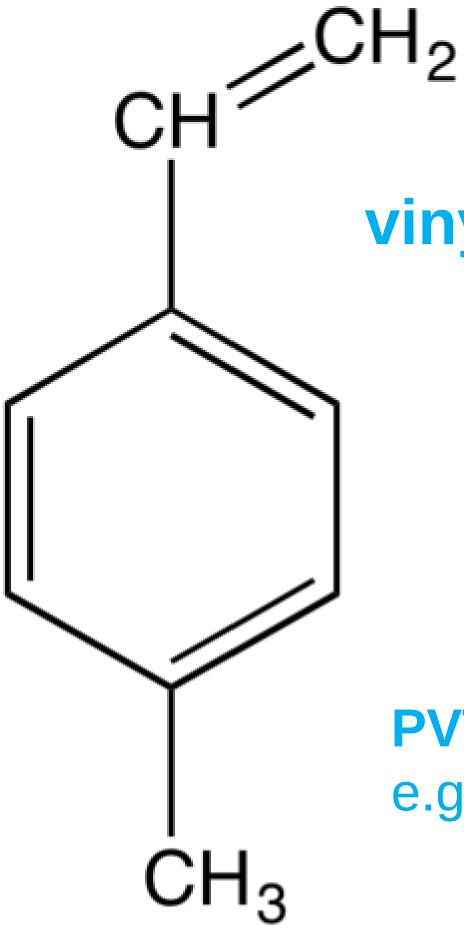
delocalized pi system

Commonly Used Polymers

styrene



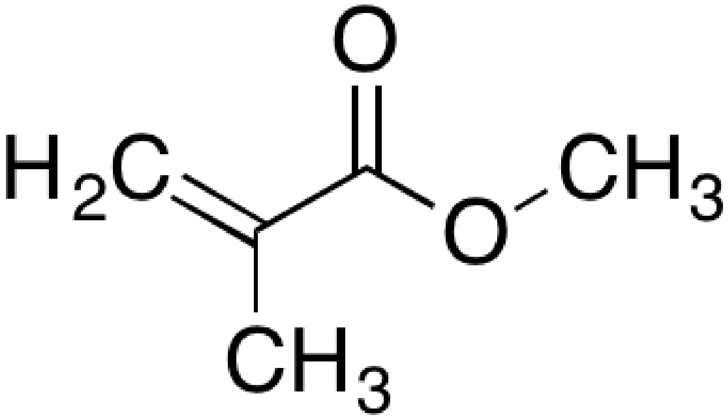
Polystyrene
e.g.: SCSN-81
CMS HCAL



vinyltoluene

PVT
e.g.: EJ-200

methylmethacrylate



PMMA
e.g.: WLS fibers

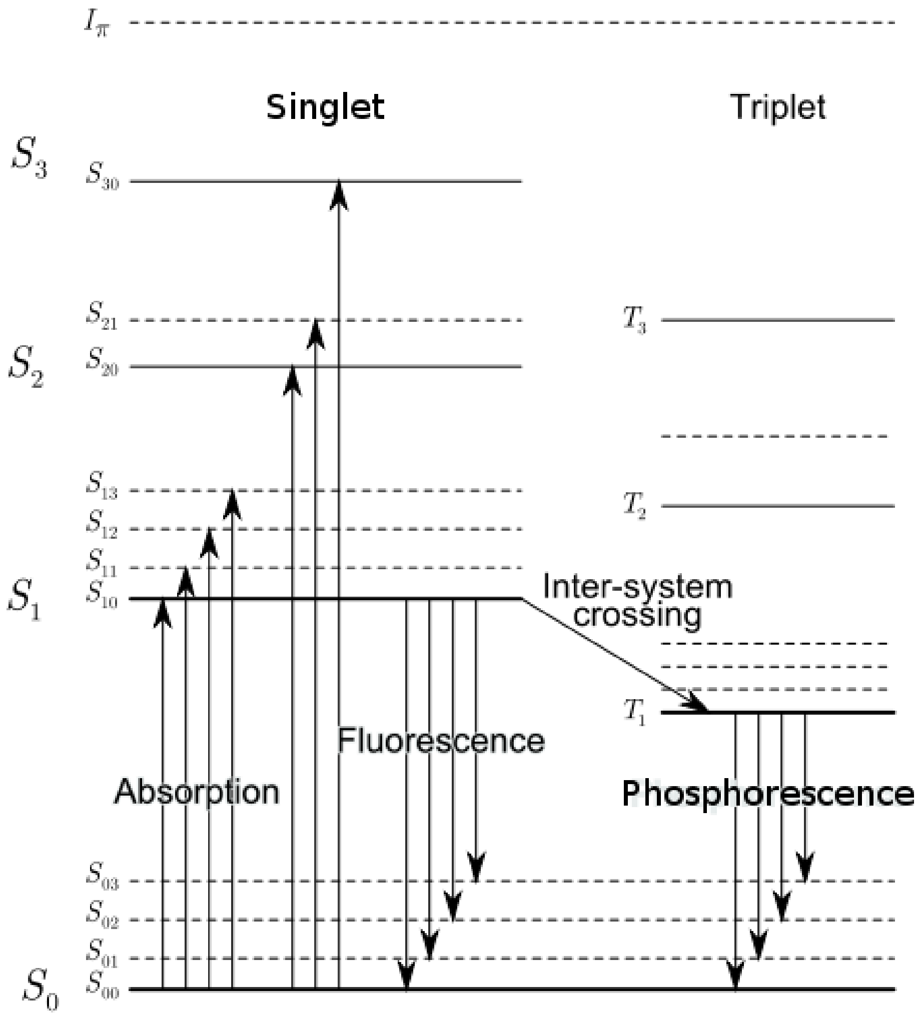
PMMA added for completeness:
not used in scintillators!

Polymer Substrate Excitation

- Four excitation mechanisms:
    1. Excitation into π-electron singlet state
    2. Ionization of π-electron
    3. Excitation of electrons other than π-electron
    4. Ionization of electrons other than π-electron

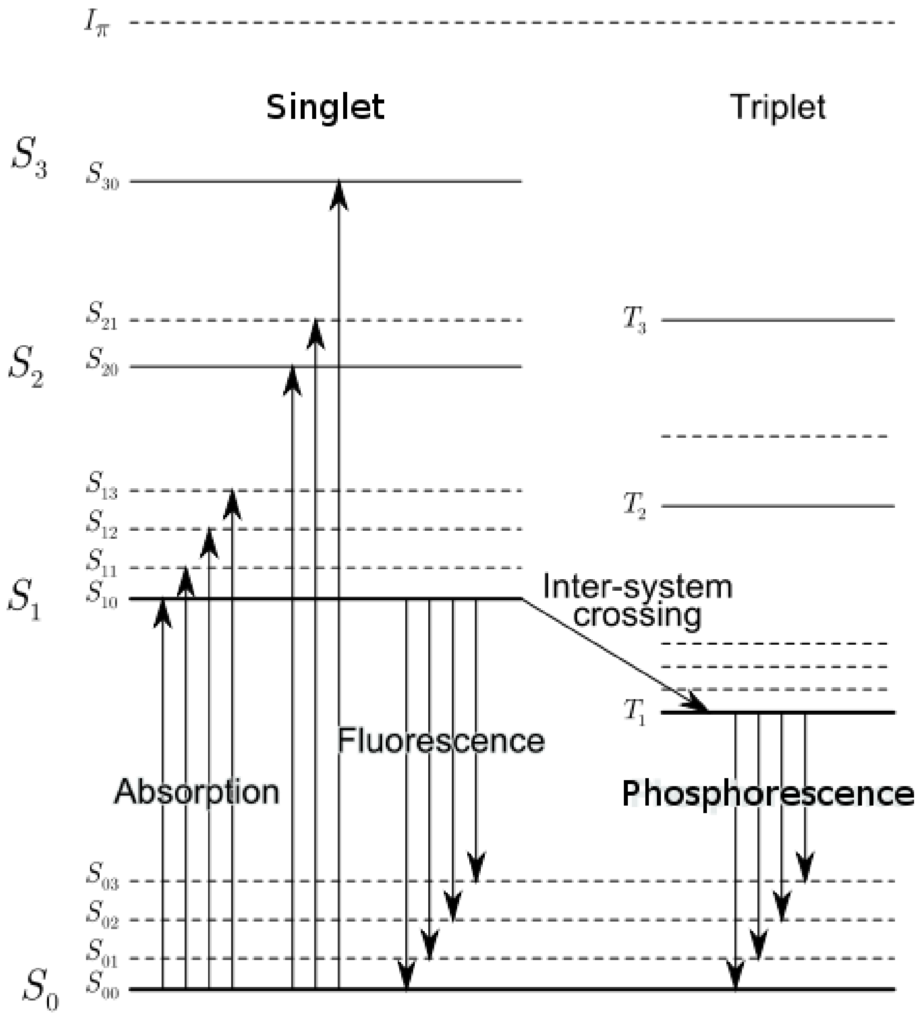
… with different outcomes:
1. Fast scintillation
2. Ion recombination leads to excited triplet or singlet π-electron states: slow scintillation
3. Thermal dissipation
4. Temporary (Birks' law) and permanent molecular damage

- Typically, 2/3 of energy yields molecular excitation, 1/3 goes to ionization
    - Scintillation probability for benzene ~ 10%
        - Multiply 2/3 by fraction of π-electrons



Light Production – Stokes' Shift

- Both ground and excited states have many vibrational sub-levels
  - Crucial feature is that inter-atomic spacing is larger in excited states than in ground states, hence de-excitation goes to sub-levels above ground S_{00}
    - Non-radiative transition to S_{00} follows
- De-excitation path leads to separation between absorption and emission spectra: Stokes' shift
  - Depends on environment around atom; how molecules are folded; proximity to other molecules; proximity of radicals



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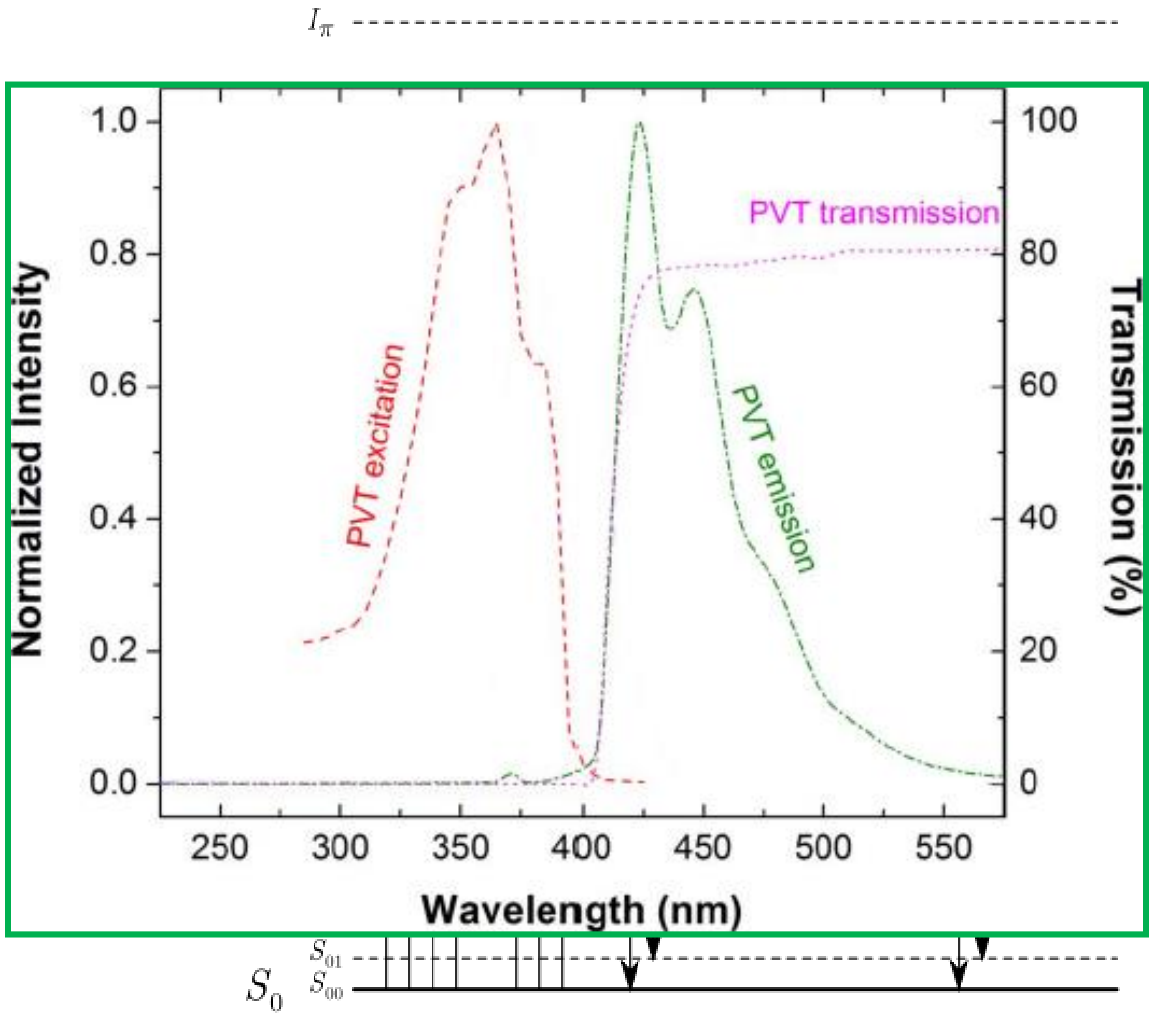
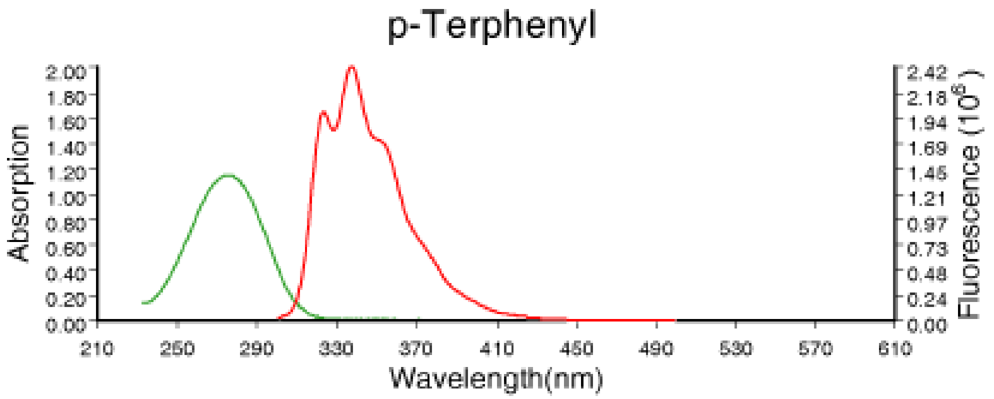
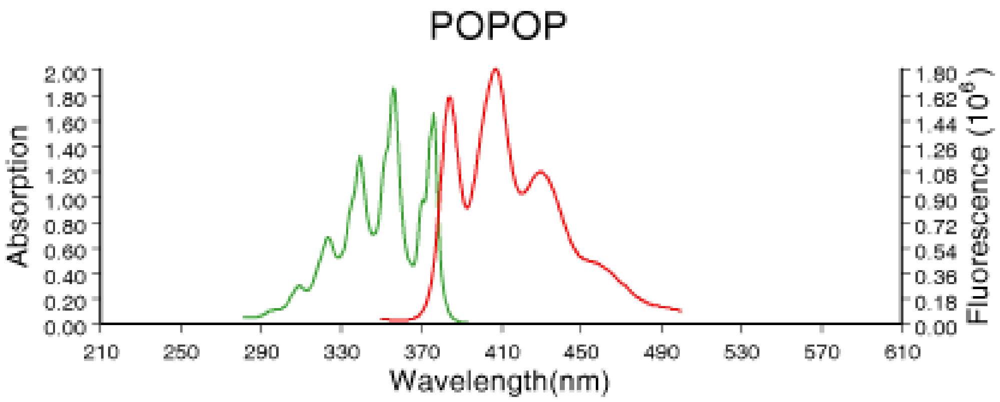


The Role of Dopants

- Energy transfer from base to primary dopant
  - Initial excitation transferred to dopants radiatively (in deep UV) or via dipole-dipole interactions (Forster mechanism)
    - Non-radiative fraction increases with dopant concentration
  - Common primary dopants: PTP (p-Terphenyl), PPO
- … and from primary to secondary dopant
  - Radiative transfer
  - Common secondary dopants: POPOP, TPB, K27, 3HF
- Executive summary
  - Dopants shift wavelength of emission further away from base-material absorption range
    - Note: Stokes' shifts *change* when dopants mixed in with base



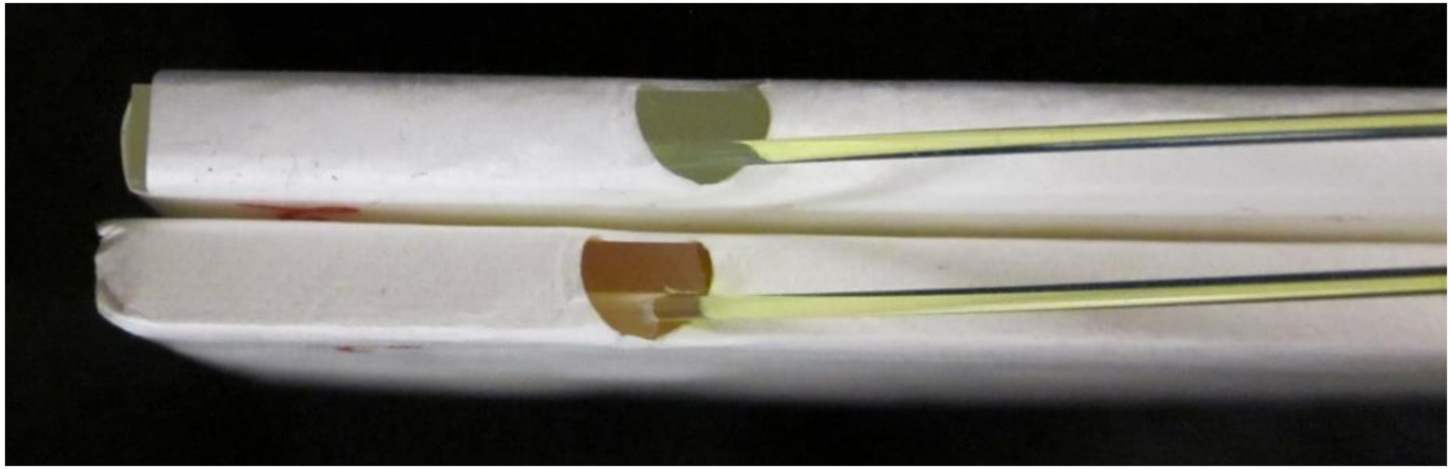


Radiation Damage

- Dominant mechanism is damage to *base material*
  - – Dopants are mostly radiation-hard
- Two components to light-yield reduction of plastic scintillator
  - – Reduction of initial light yield
  - – Absorption of light produced by secondary dopant
    - “Color centers” reduce the attenuation length

Effects of radiation:

- Breaks polymer chains and create radicals that absorb UV light
  - Irradiated scintillator turns dark

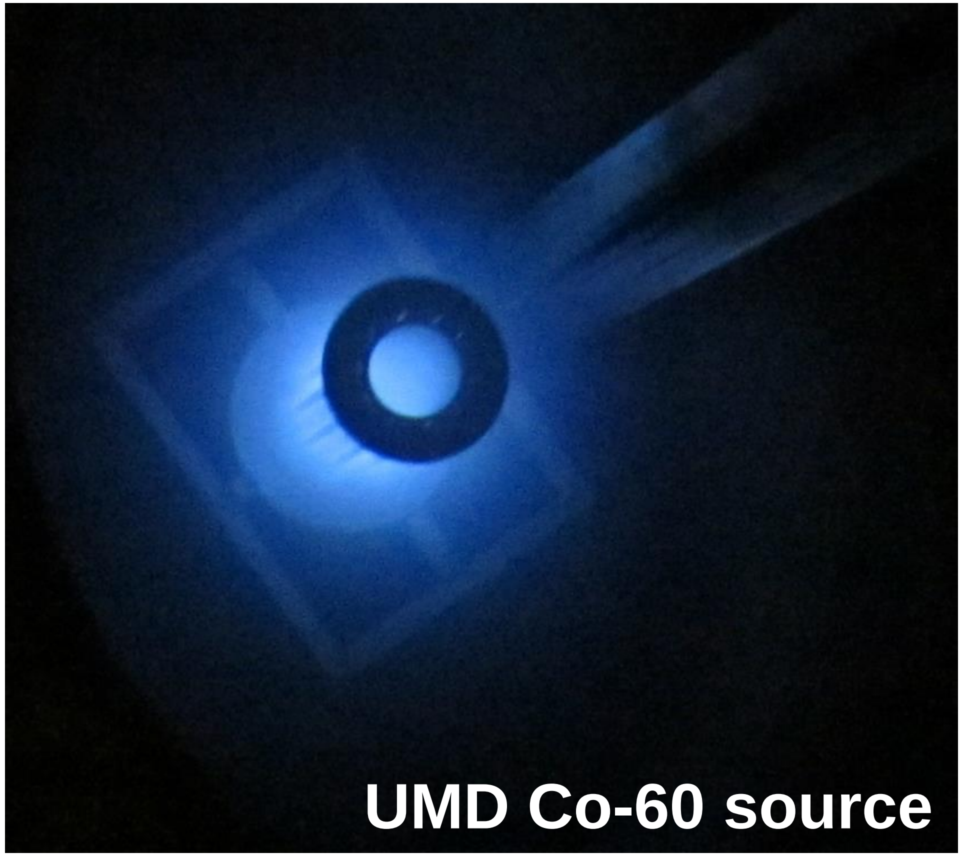


Some parameters to model radiation damage

- Presence of oxygen
- Total irradiation dose and dose rate
- Temperature of irradiation

Investigating Radiation Tolerance

- Identify candidate materials offering improved radiation tolerance
  - Tune dopant concentration
  - Emit at a longer wavelength
- Irradiate materials in different environmental conditions, at different total doses and dose rates
  - Radioactive sources (Co-60, Cs-137)
  - LHC beam halo: CASTOR Radiation Facility
- Measure light yield with different and complementary methods
  - Spectrofluorometers, cosmic rays, radioactive sources
- Map light-yield reduction as a function of multiple parameters
  - O_2 concentration; total dose; dose rate; temperature; dopant concentration…

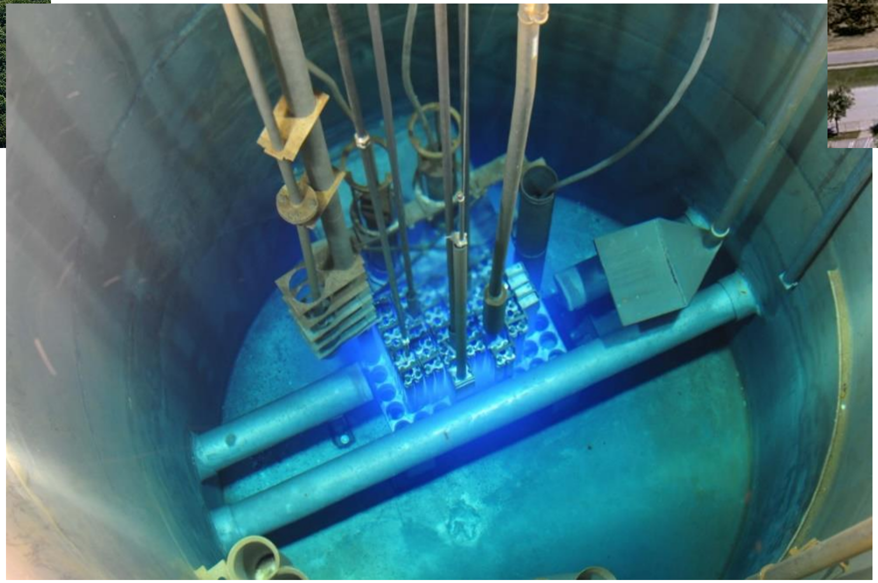


Irradiation Facilities (1)

University of Maryland

- Co-60 source
- 50-1500krad/hr

(picture: TRIGA reactor…)



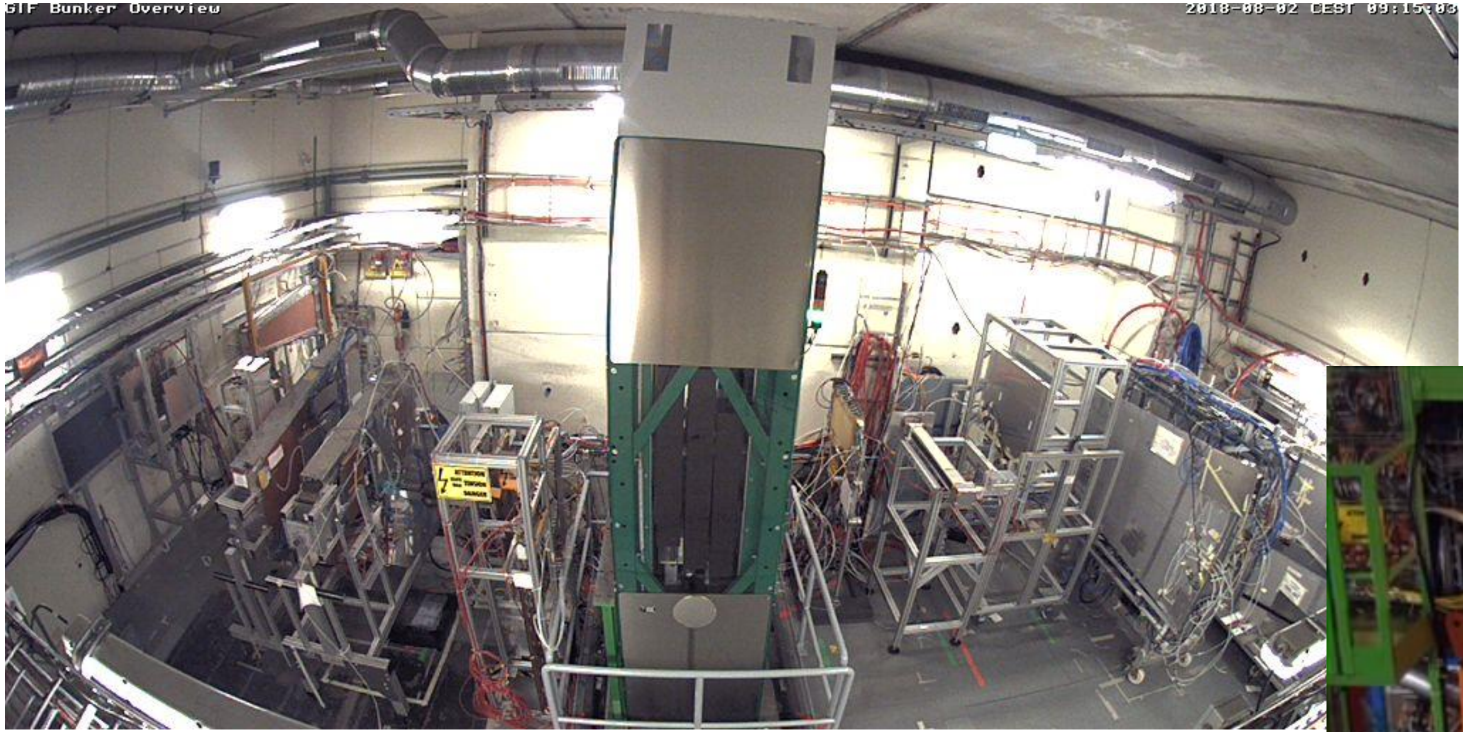
Goddard Space Flight Center

- Co-60 source
- 0.3-100krad/hr
- Cold (-30C) and warm irradiations

NIST

- Co-60 source
- 50-500krad/hr
- Cold (-30C) and warm irradiations

Irradiation Facilities (2)

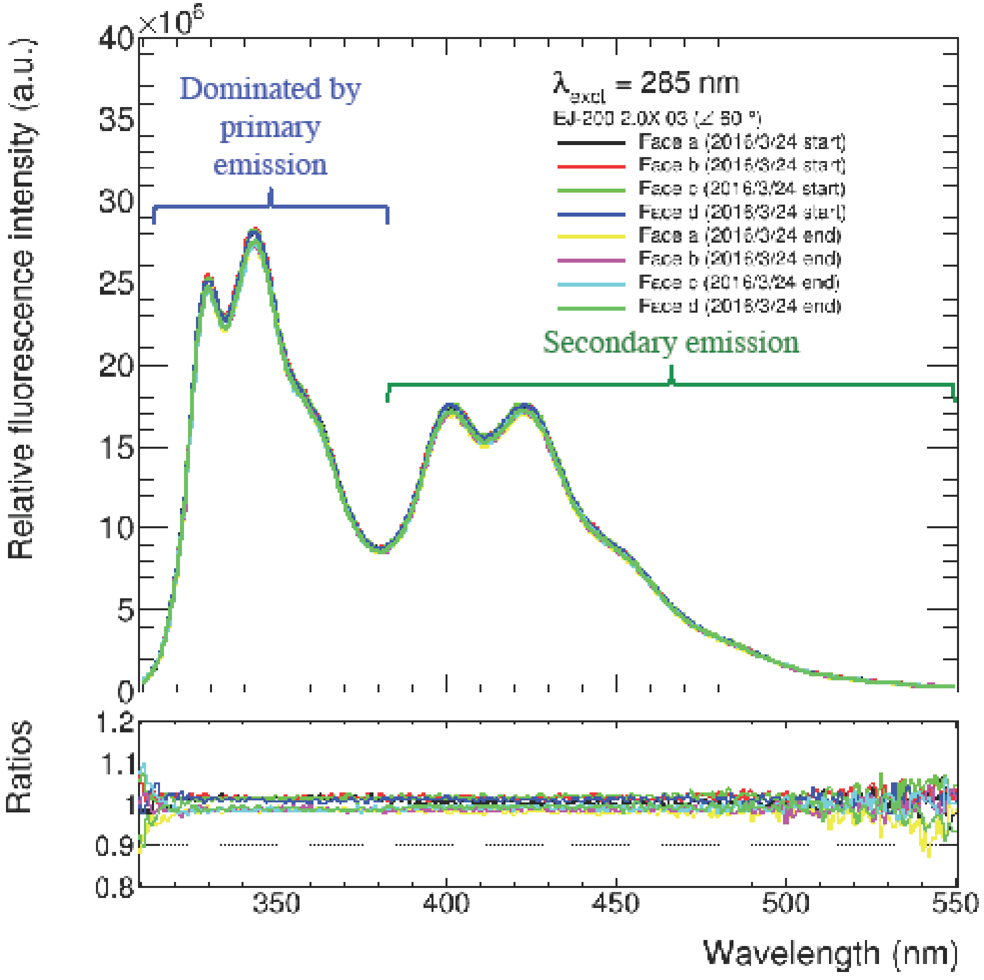


CERN CASTOR Calorimeter Table
- LHC environment
- O(10) of CMS highest dose rate

CERN GIF++
- Cs-137 source
- 0.05krad/hr

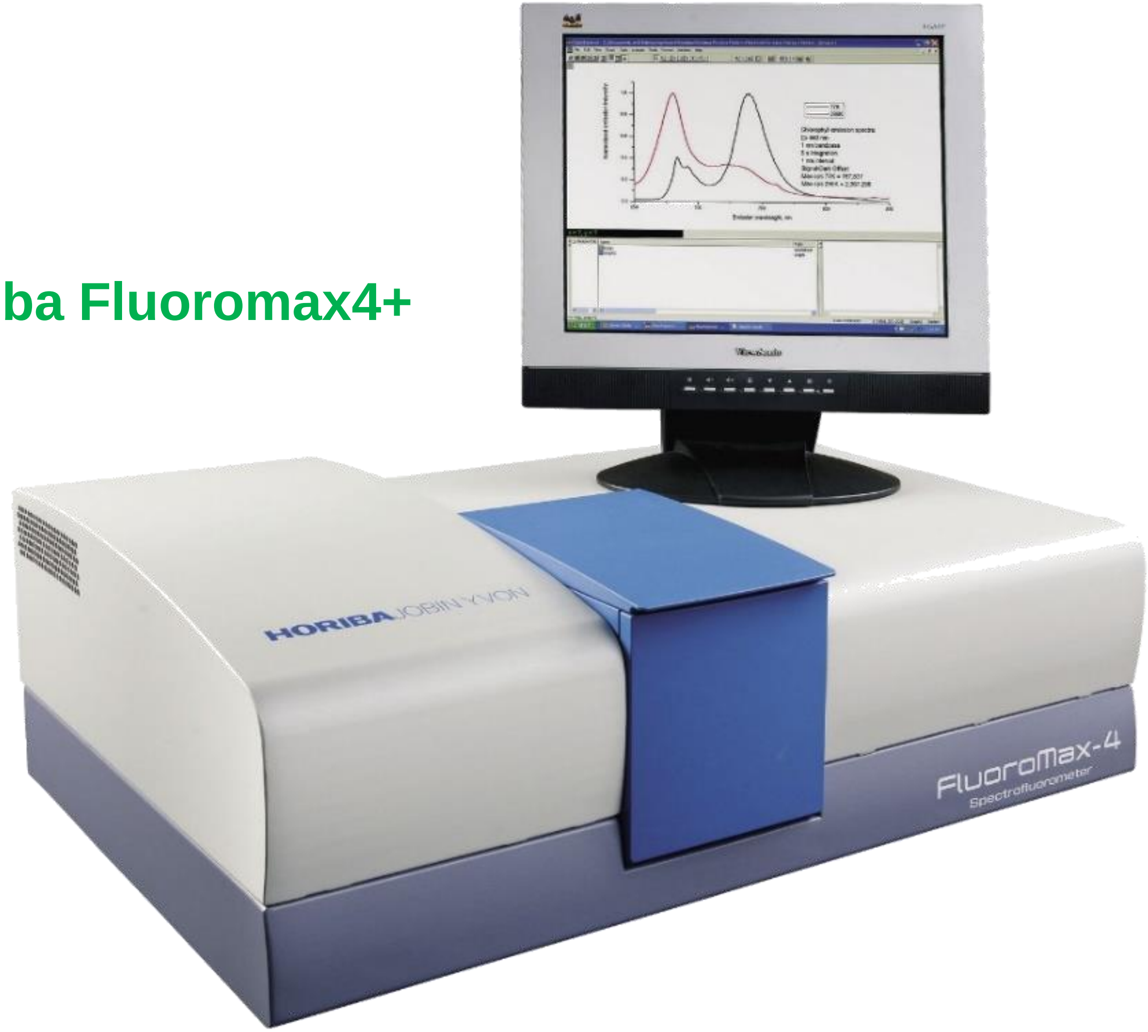
Spectrofluorometry (1)

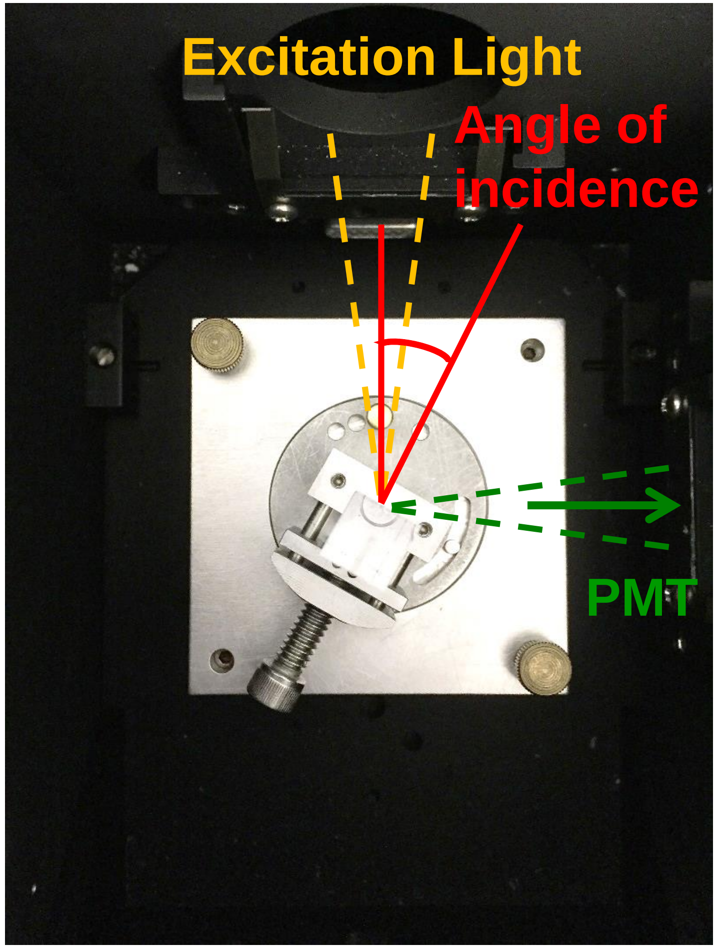
- Very challenging measurement
  - Typical user needs accurate measurement of peak positions, not peak amplitude
- Tuned procedure until reached satisfactory level of repeatability
  - Repeated measurements during a day vary within <2%
    - Include uncertainty on machine conditions, placement of sample by operator, inhomogeneity among sample sides
- Possible to probe effect of radiation on dopants separately by varying excitation wavelength
  - E.g. blue scintillator: 285nm (excite primary), 350nm (cross primary/secondary), 400nm (excite exclusively secondary)



Spectrofluorometry (2)

Horiba Fluoromax4+

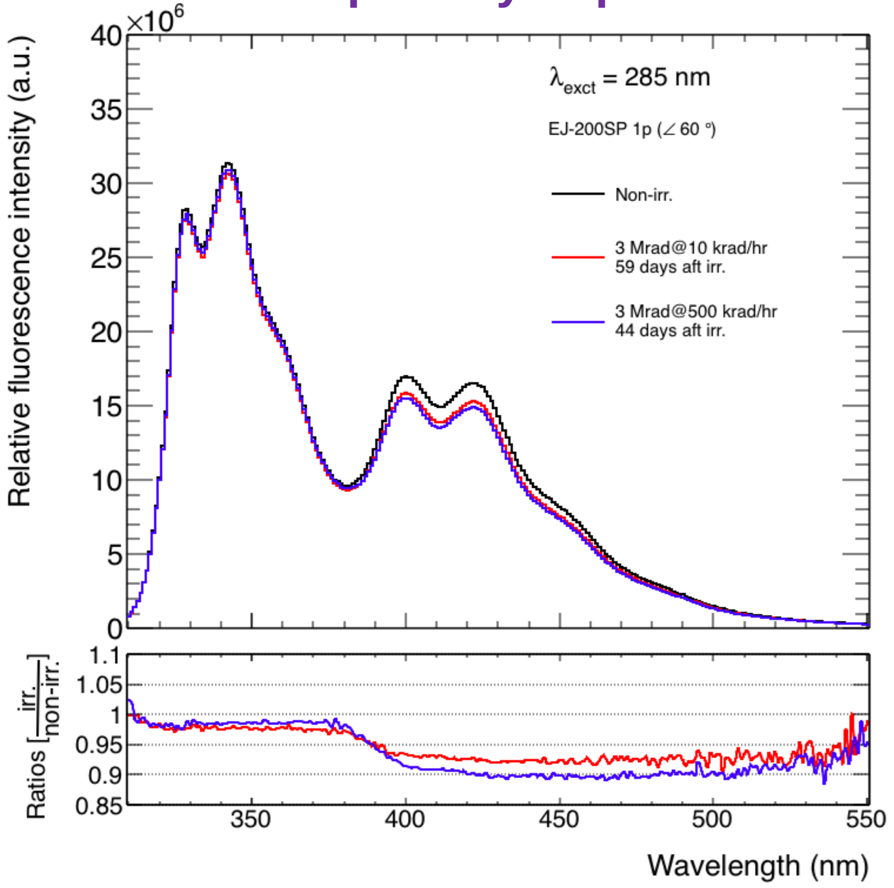




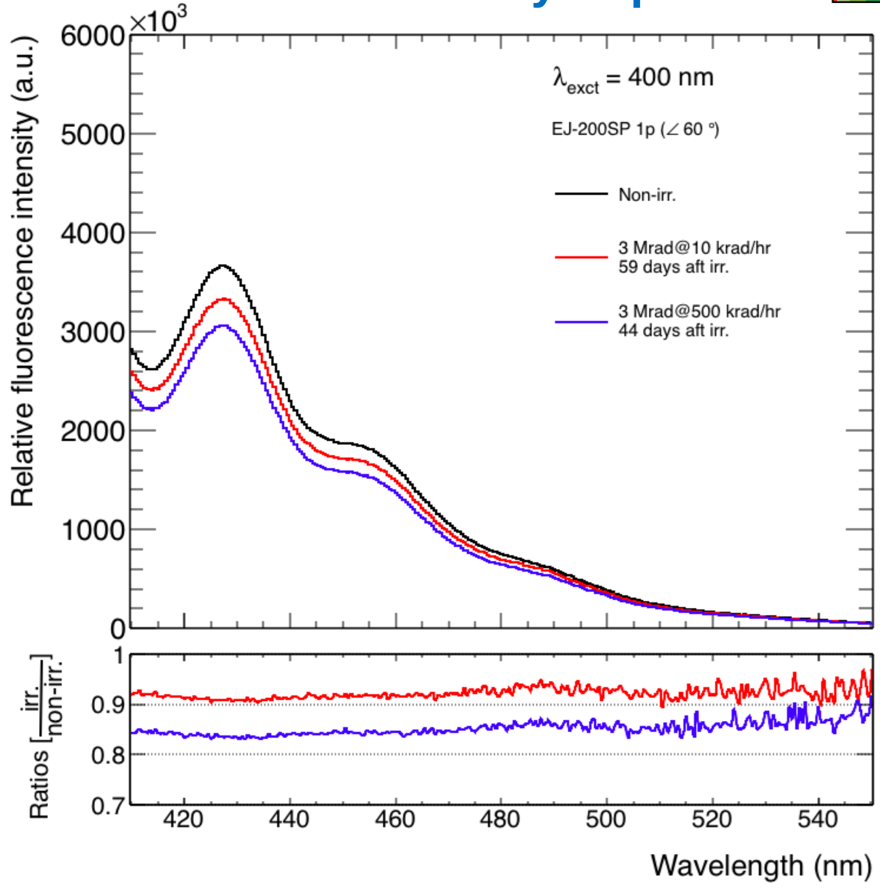
UMD-designed sample holder

Spectrofluorometry (3)

Excite primary dopant



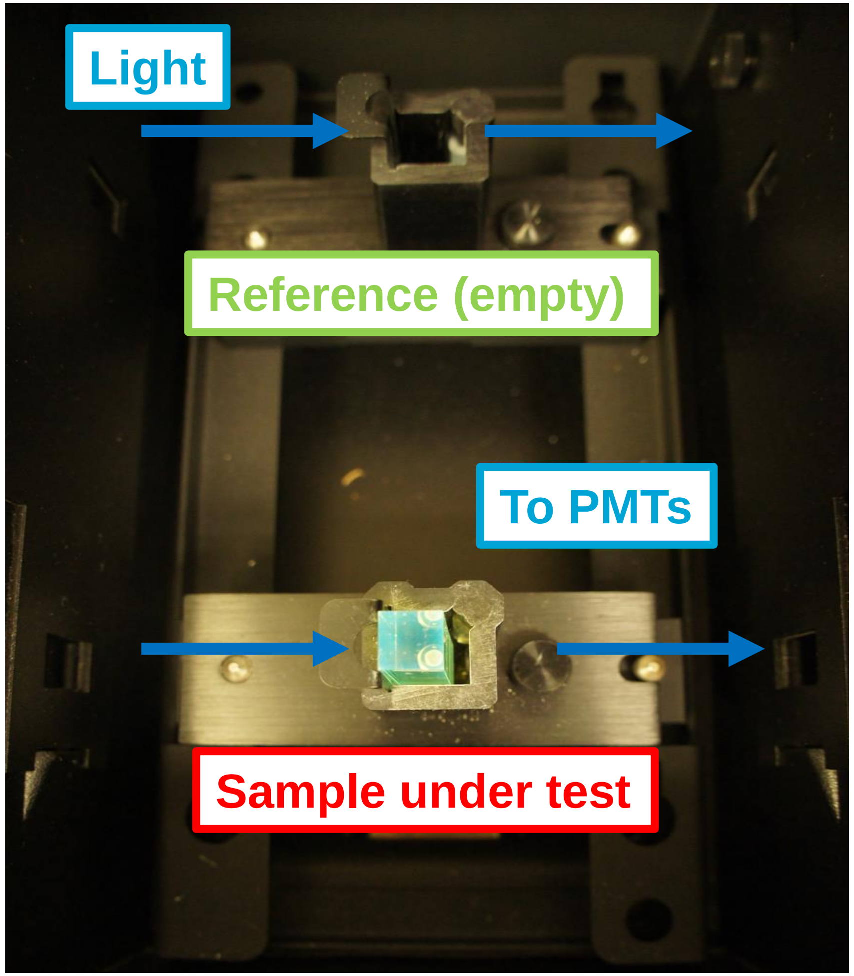
Excite secondary dopant



- Technique allows one to understand effect of radiation on dopants
  - One can excite dopants separately, and check efficiency of energy transfer between them

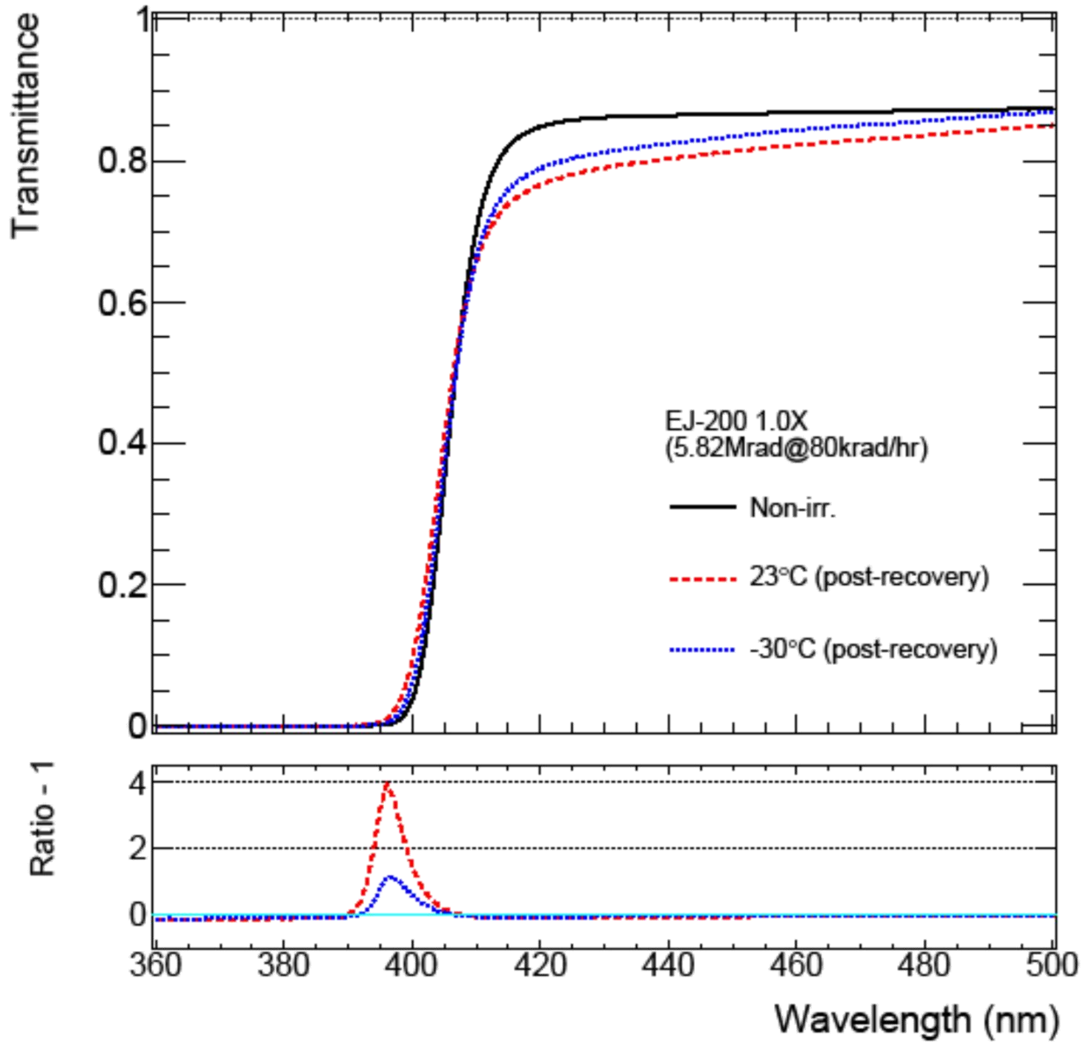
Transmission/Absorption

- CARY 300 UV-Visible spectrophotometer
  - Double-beam mode to reduce uncertainties
- Measurement (somewhat) sensitive to bulk effects
  - Samples are 1-cm thick, completely traversed by incident light
  - Measure annealing times of order ~ few weeks
- Absorption spectra used as input to GEANT simulations
  - Important step in understanding plastic damage is availability of tuned simulation of optical properties of plastic



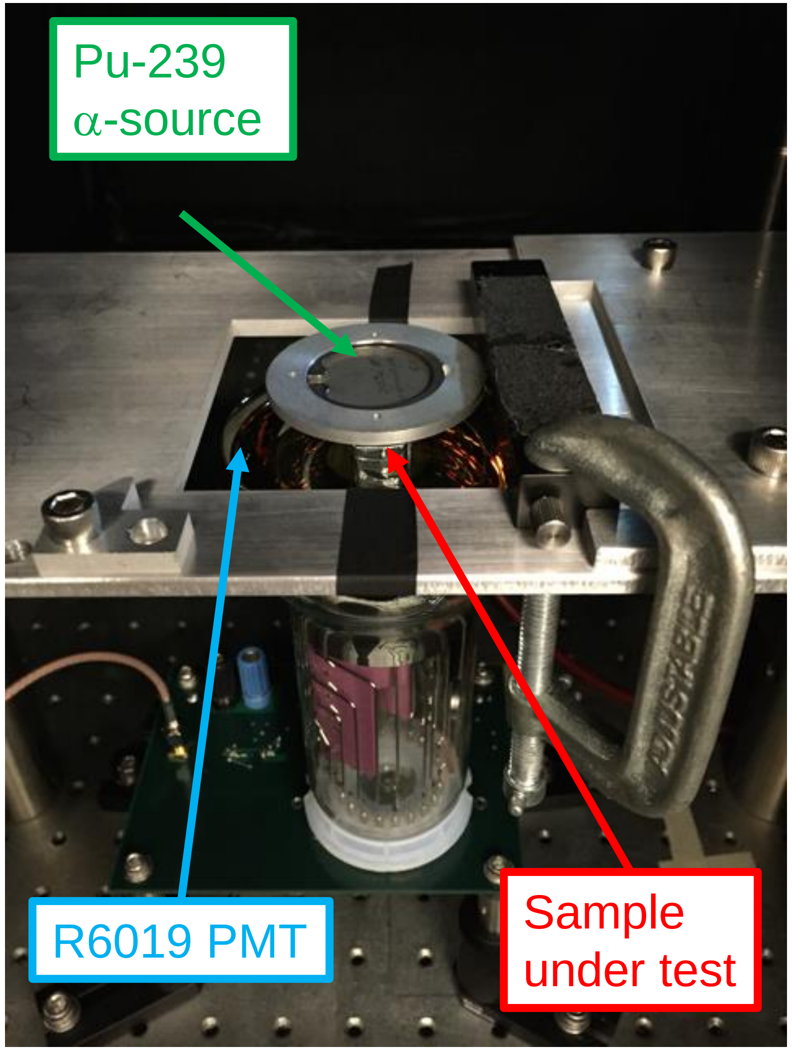
Transmittance Measurements

- Measurement details
  - Commercial EJ-200
  - 5.82Mrad at 80krad/hr, NIST
  - Irradiation at 23C vs. -30C
  - Samples annealed about 20 weeks at room temperature
- Observations
  - Peak at ~400nm (absorption maximum of secondary dopant) seems to indicate some damage of secondary dopants
    - Less dopant to absorb light → higher transmittance
  - Comparable transmittance above 410nm after annealing



Alpha Source Measurements

- Sensitive to *complete* chain of light production
  - Source releases energy in the base, and the whole chain of dopants and energy transfers is exercised
    - Spectrophotometer cannot produce UV light to mimic base-to-primary transfer
  - Somewhat sensitive to bulk damage
    - Energy released at small depth; light transverses about 1cm of scintillator to reach PMT
- Provides complementary measurement to transmission and emission spectra
  - Closer to actual operation of scintillator in detector



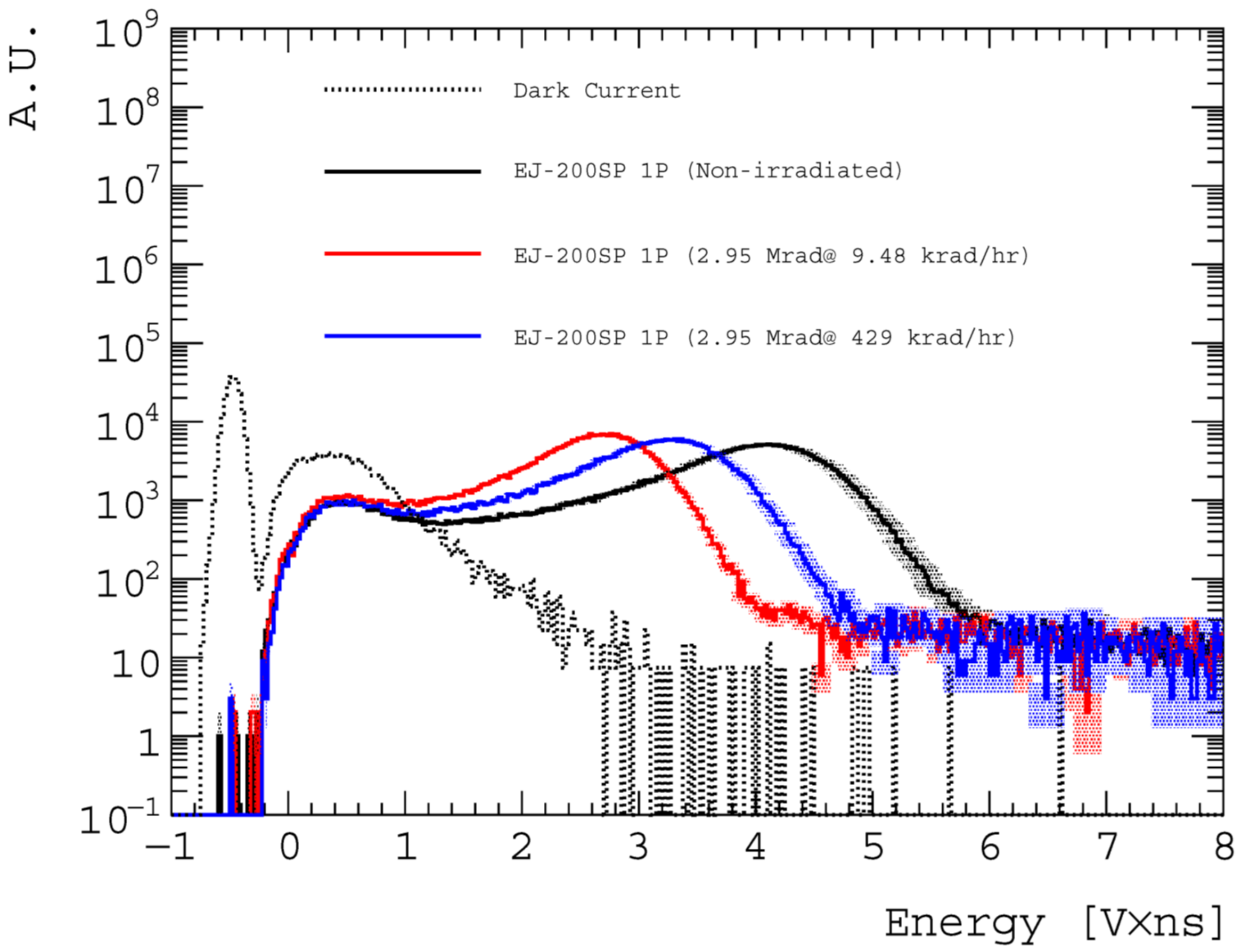
Dose-rate Effect (1)

- A counter-intuitive re-discovery
  - When the same dose is integrated over a longer period, the damage is larger
- First reports of dose-rate dependency in ’90
  - Working hypothesis: oxygen diffusion into plastic permits more reactions that create UV-absorbing radicals
- Light yield decreases exponentially as a function of integrated dose *d*:

$$L(d) \propto e^{-\frac{d}{D}}$$

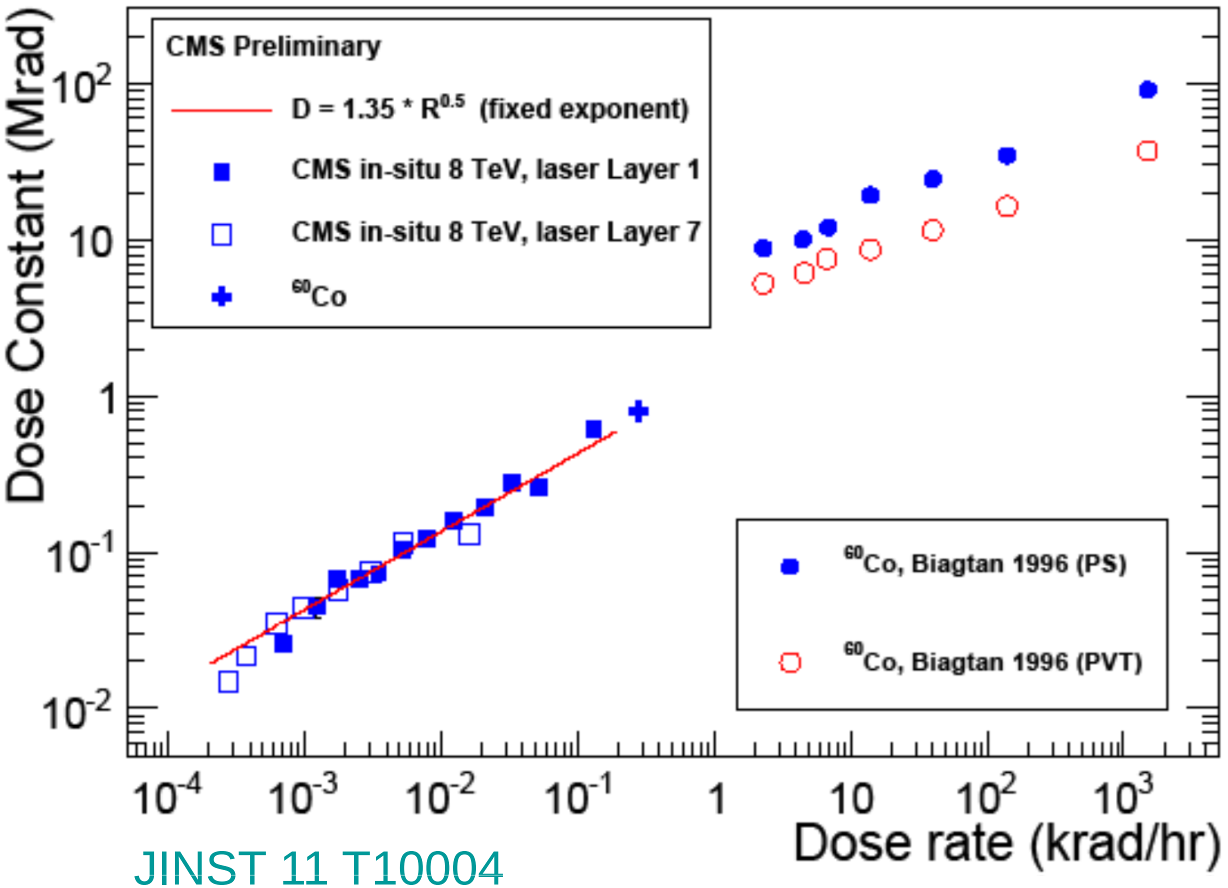
  - The *dose constant D* increases as the dose rate does

Hatched area: systematic uncertainty



Dose-rate Effect (2)

L(d) = L(0)·exp(-d/D); d: dose, D: dose constant



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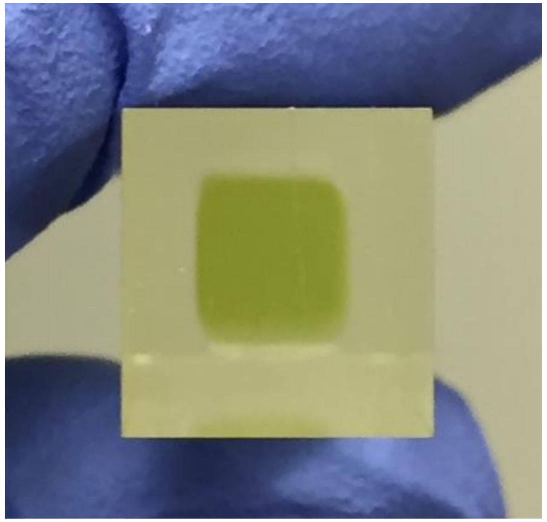
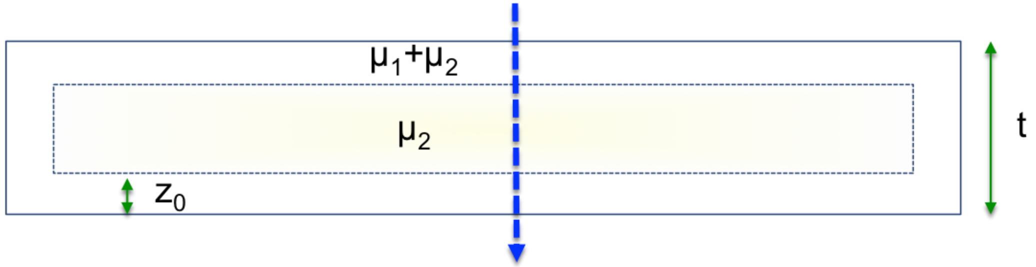
- Radiation damage (per unit of integrated dose) increases at low dose rates
  - Power law between dose constant and dose rate matches what we would expect under the assumption that oxygen diffusion drives the dose-rate effect
    - Is oxygen diffusion driving the dose-rate dependency?



More to Dose-rate Effect

- Oxygen diffuses up to a depth z_0 into the substrate
  - Diffusion depth proportional to $^1/_{\sqrt{R}}$, where R is the dose rate
  - Proportionality coefficient depends on diffusion constant, solubility constant, oxygen pressure, and rate of formation of radicals
- The absorption coefficient models the light output
  - Defined as the product of the density of color centers and their cross section for light absorption
  - The color-center density and type depend on the presence of oxygen
- The light yield can be written using a dose-rate-dependent effective absorption coefficient
  - Dose constant $D = \frac{\sqrt{R}}{a+b\sqrt{R}}$
  - Observe $\sqrt{R}$ dependence of dose constant for small dose rates; expect D to tend to a constant value for high dose rates (oxygen has no time to diffuse at all)

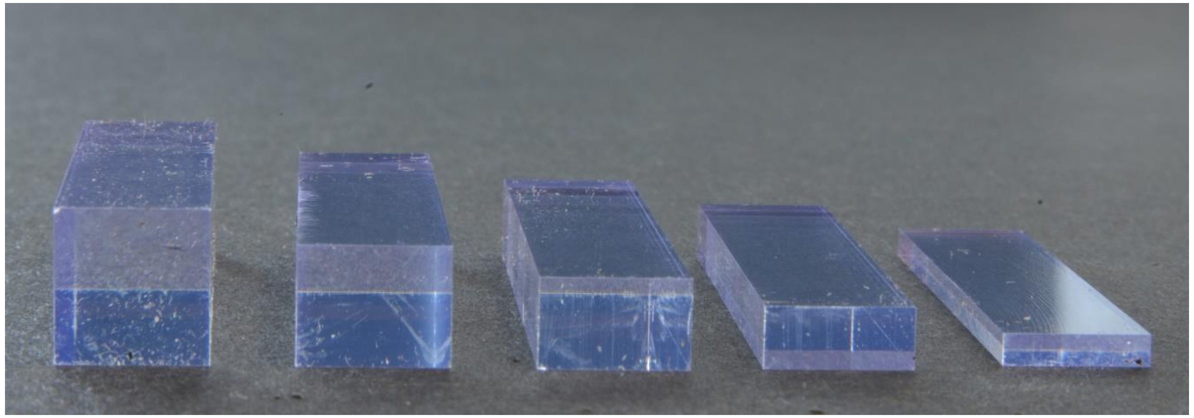


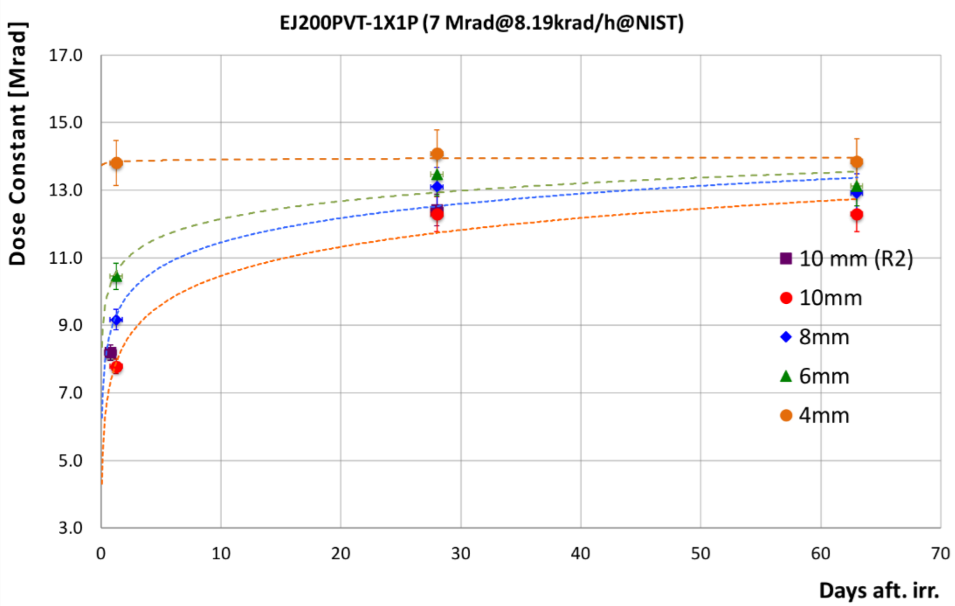
$$L \propto e^{-\mu_1 \cdot 2z_o - \mu_2 \cdot t}$$

t : sample thickness
z_0: oxygen diffusion depth
μ_1: absorption coefficient in the presence of oxygen
μ_2: absorption coefficient independent of oxygen

Variant Thickness Studies

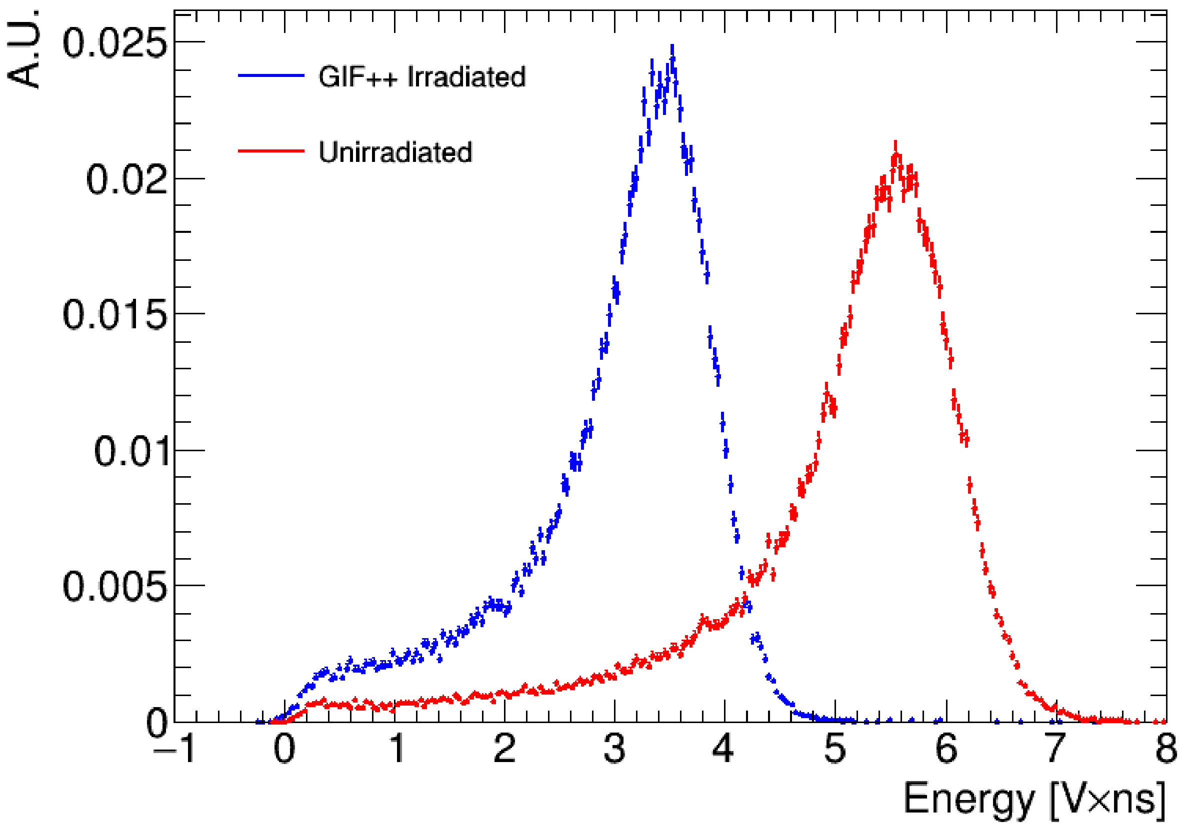
- Attempt at disentangling effect of oxygen diffusion on absorption coefficient
  - Measure effective absorption coefficient in lab using transmission/absorption and α-source measurements
- Laboratory measurements of samples with different thickness used as inputs to GEANT4 simulation
  - Final goal is measurement of wavelength-dependent absorption coefficients in oxygen-depleted vs oxygen-filled regions, and of diffusion depth z_0 vs radiation dose rate





Very-Low Dose-Rate Studies

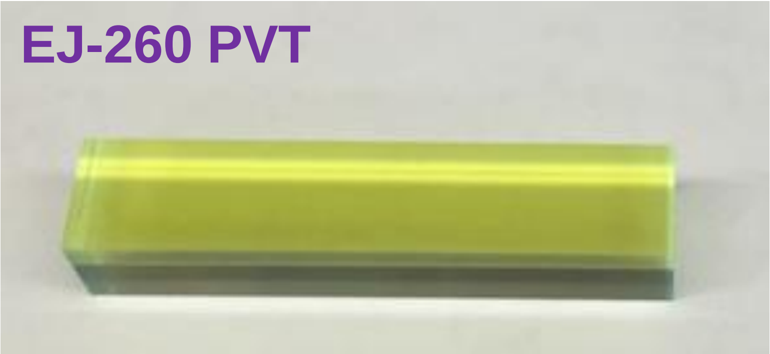
- Lowest dose-rate measurement performed in-situ with HE laser and radioactive-source calibration system
  - First results presented after integration of 0.2Mrad in about two years of LHC operations (2010-2012)
    - Continued to update results as more data were collected
  - Radiation effects on scintillator, wavelength-shifting fibers, and photosensors are combined
- GIF++ facility allows for probing similar dose rate as in the case of the HE detector
  - Cs-137 source, dose rate ~ 50rad/hr
  - Irradiated samples measured in laboratory, and radiation damage on plastic measured independently of other contributions



2-year long irradiation at GIF++
~300kRad @ 50rad/hr
α-source measurement

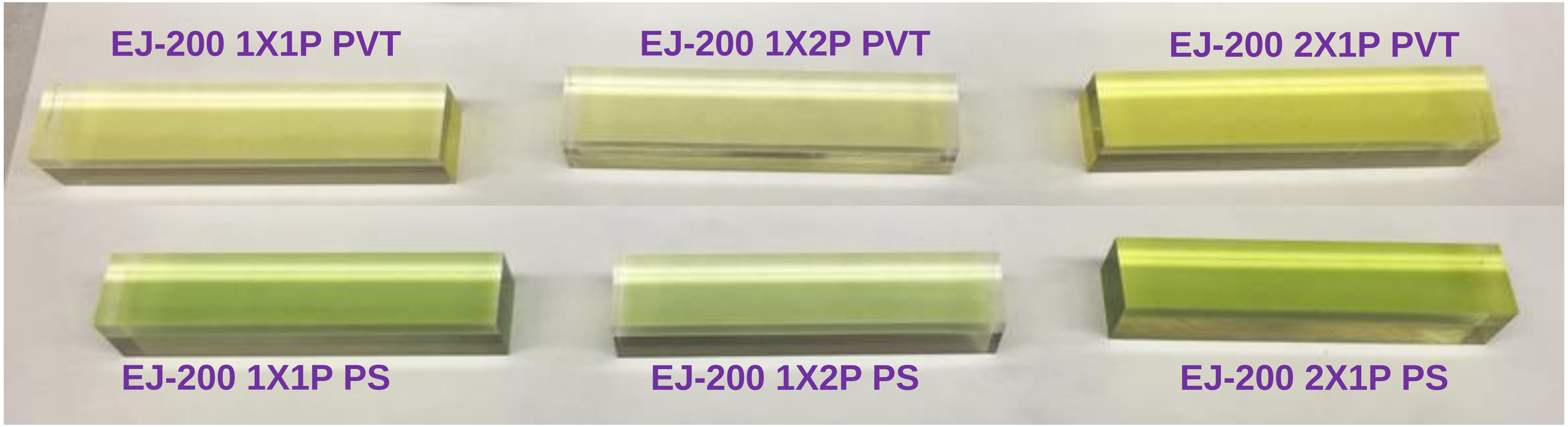
Base-Material Studies

- Investigation of scintillator produced with same dopant configuration, and different base
  - Green and blue fluors
  - Normal concentration of fluors; over-doped primary (2x); over-doped secondary (2x)
  - Polyvinyltoluene and polystyrene base
- CMS Hadron Calorimeter uses PS-based scintillator; current commercial scintillators mostly PVT-based
  - One note of interest: oxygen diffusion coefficient (measured in cm^2/s) is 13 times larger in PVT than in PS
- Measurements on irradiated samples suggest that PVT-based scintillators are more radiation-tolerant than PS-based scintillators



Co-60 Irradiation at NIST
7Mrad @ 500krad/hr

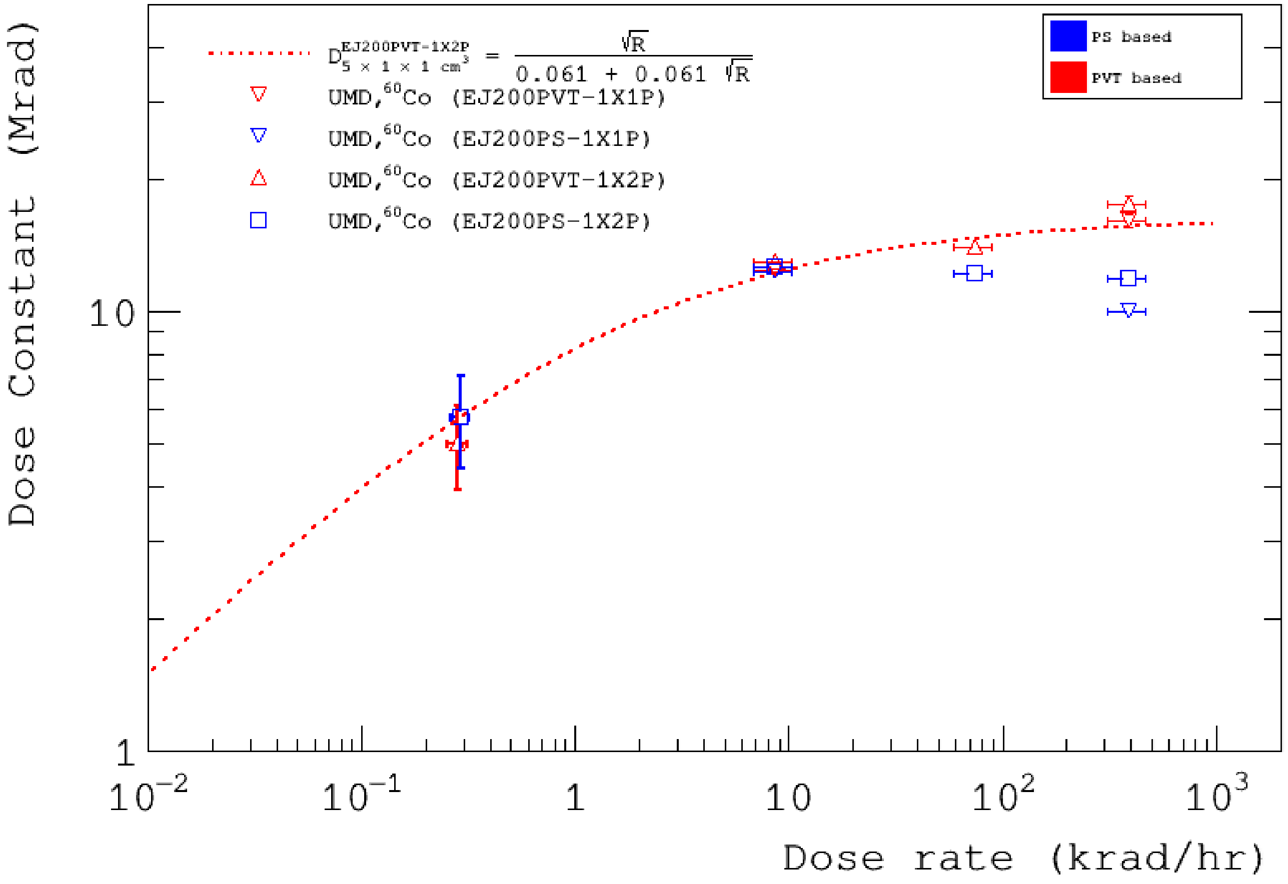
Over-doping Studies



- Polystyrene vs polyvinyltoluene blue scintillators; Co-60 irradiation, 7Mrad @ 500krad/hr
  - 1X1P: commercial version; 1X2P and 2X1P: over-doped versions
    - The concentration of the primary or secondary dopant is doubled
- Pictures suggest that over-doping helps preserve the scintillator clear, and confirm that PVT seems to hold better than PS
  - Measurement of dose constant reveals that over-doping marginally improves radiation tolerance
    - Important note: the 1X1P and 1X2P samples annealed for about 12 hours longer than the 2X1P

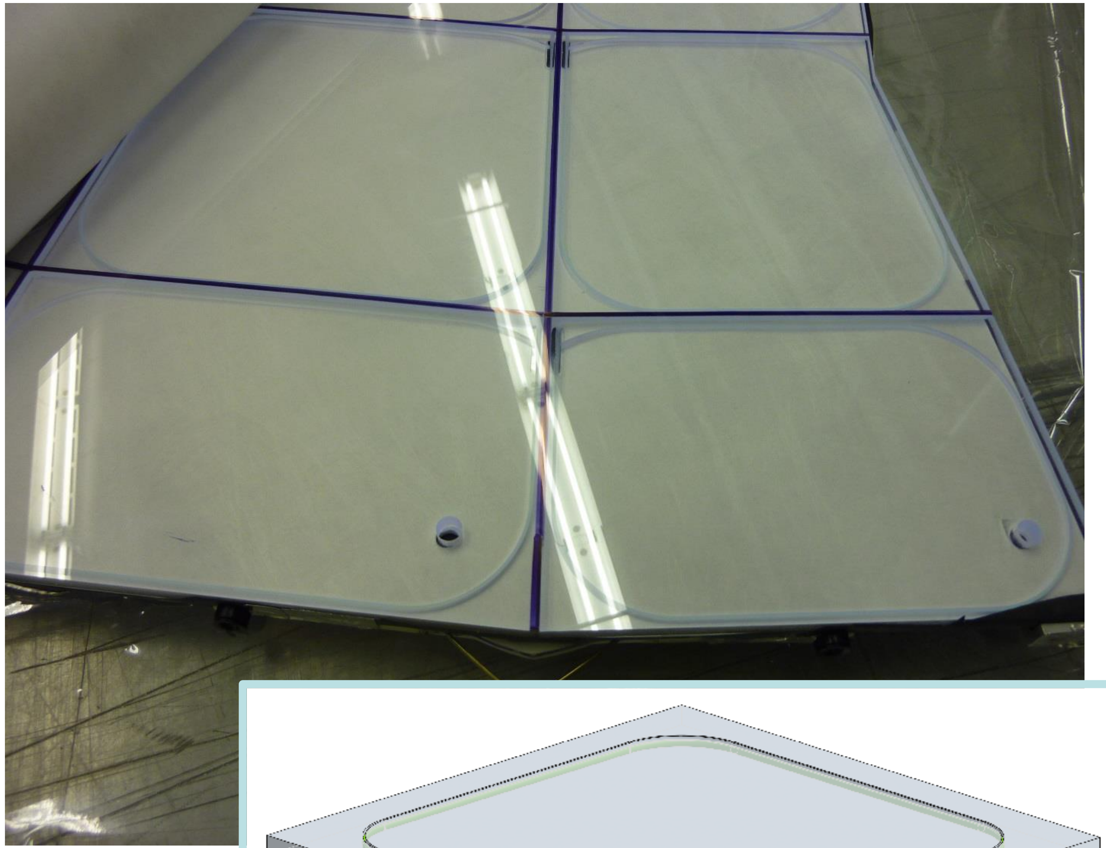
Dose-Constant Summary

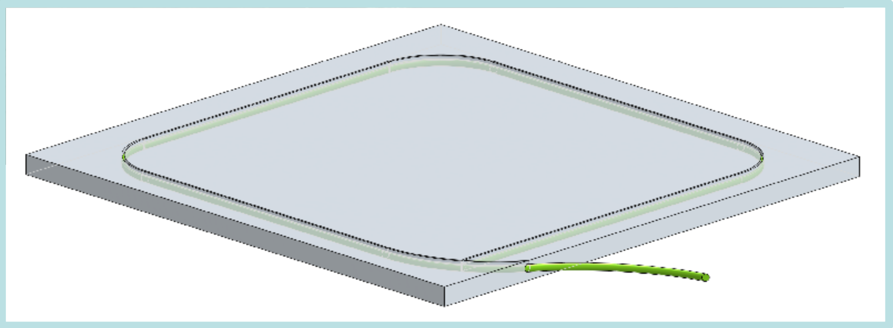
- Basic model captures behavior of plastic scintillator under irradiation in large range of dose rates
  - Ideally, would need more low-dose rates to check behavior
- Quick take-home message from plot
  - PVT performs better than PS
  - Over-doping improves radiation tolerance marginally
- More measurements available
  - Some left out to avoid cluttering the plot, some need to be cross checked



CMS HCAL σ-Tiles

- Studies on scintillator properties performed using $1x1x5cm^3$ rods
  - Mechanical constraints imposed by spectrophotometers
- Tiles in CMS HCAL are thin squares, with a wavelength-shifting fiber inserted in a groove close to the edge
  - Typical size: $10x10x0.4cm^3$
  - The σ-tile design demonstrated to maximize uniformity of light collection vs. particle crossing position
- The light collected by the WLS fiber is then transported to photosensors via a clear fiber
  - Setup allows photosensors to be installed in an area with lower radiation



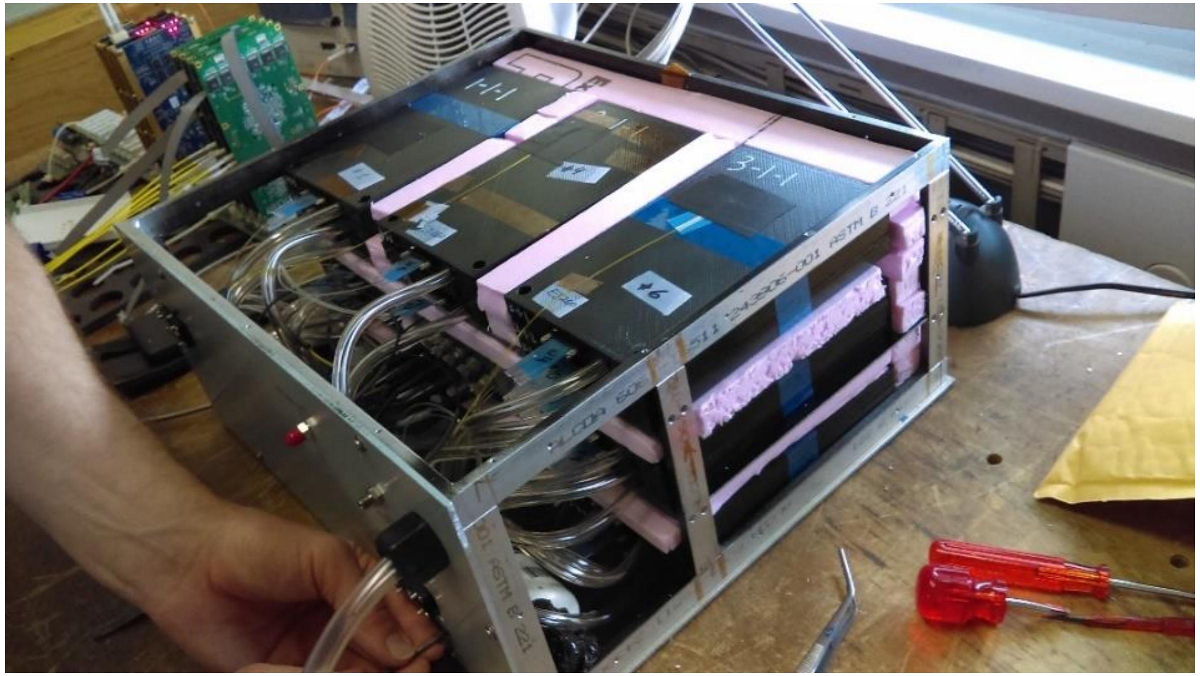


Irradiations @ LHC

- Comprehensive set of samples installed next to LHC beam
  - From scintillator in current HE to patent-pending materials
- Each tile is connected to the CMS DAQ and HCAL Calibration systems
  - A laser fiber can excite directly each tile, and provide a signal with known amplitude
- The system allows for the continuous monitoring of scintillator ageing
  - Irradiation conditions more closely match the conditions of actual detectors

Castor Radiation Facility

- Two rounds of irradiations, in different position w.r.t. LHC beamline
  - Investigated different range of dose rate
- Very challenging effort compared to laboratory measurements
  - Light collected via wavelength-shifting fiber connected to clear fiber
  - Photosensors also installed in radiation area
- Ongoing analysis of live data collected during LHC operations
  - One-time measurement of scintillator performance in laboratory (after annealing) useful to normalize results



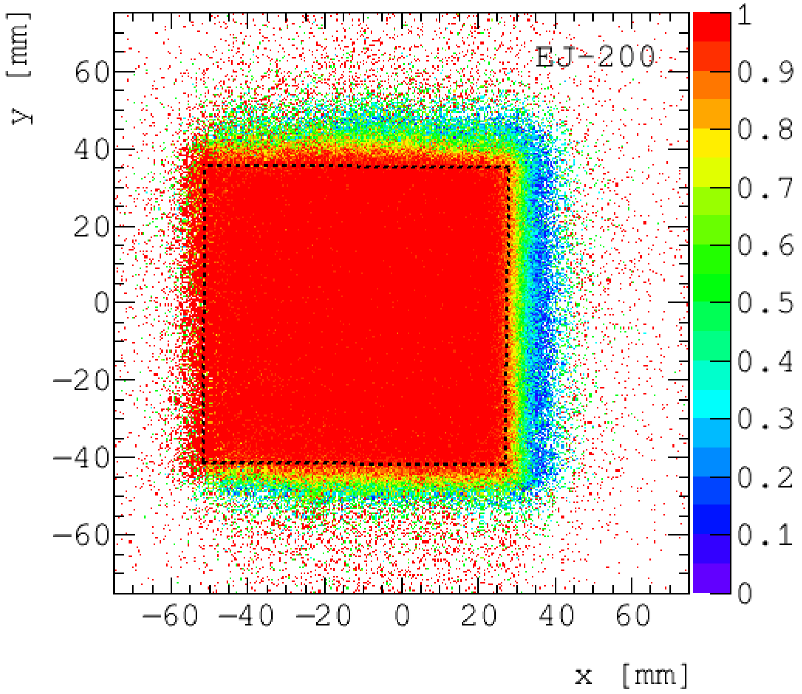
CRF Scintillator boxes

Tile Testing at CERN H2

- Test beam facility at CERN
  - Focused on 150GeV muon sample (MIP)
  - Tracking information provided by set of wire chambers
- Sample tiles connected to full CMS HCAL DAQ chain
  - Test of both the scintillator and the data-acquisition system
- Measured light-collection efficiency and yield
  - A collection of unirradiated scintillator samples

CMS HCAL Wedge

JINST 13 P01002

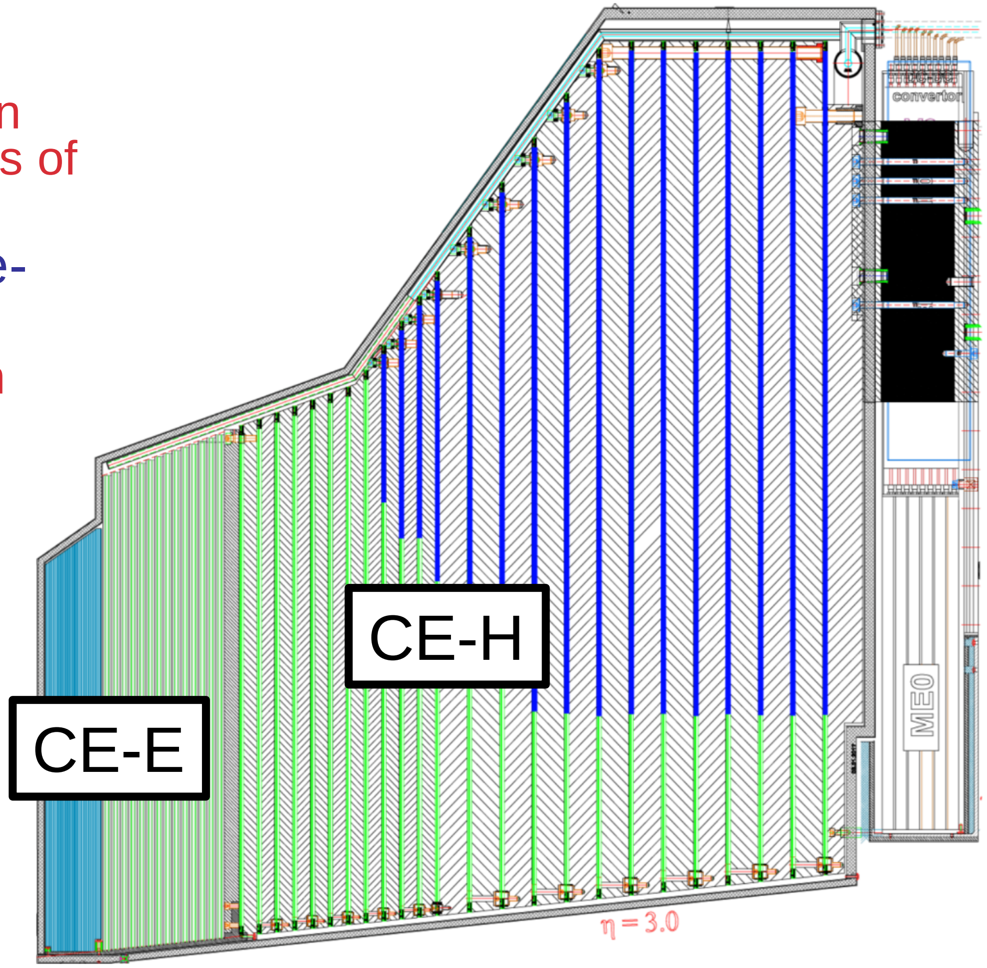


EJ-200: hit efficiency map

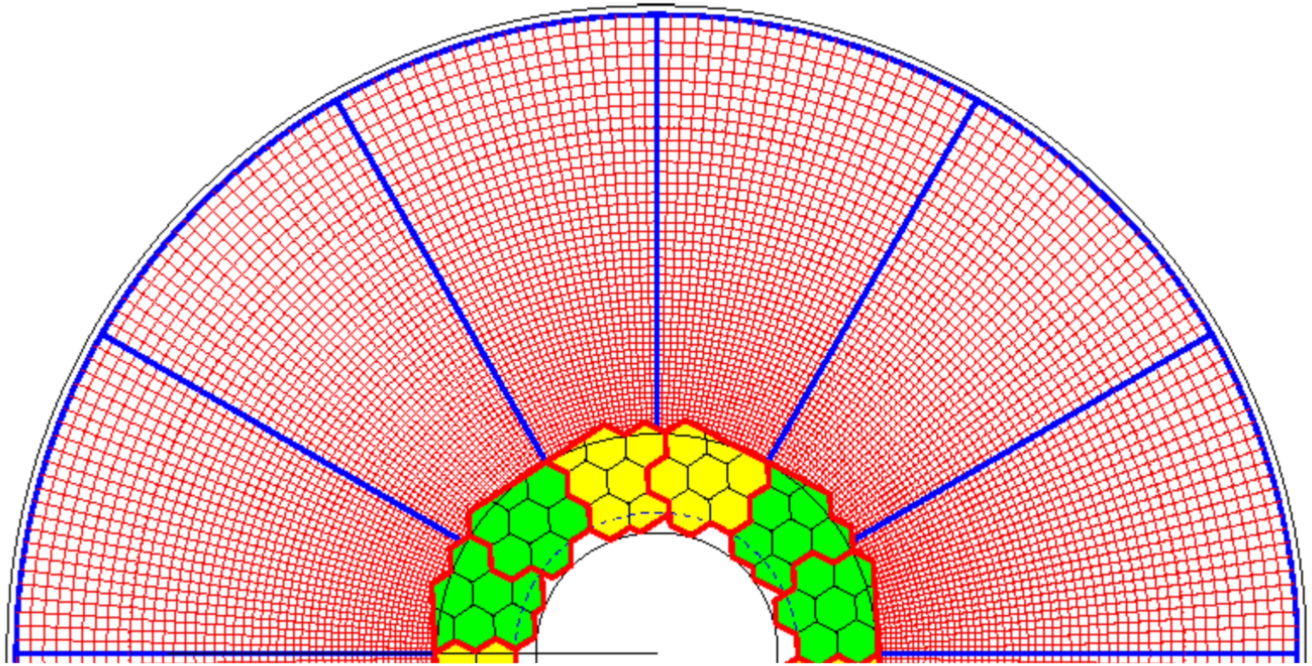
On-going measurement of uniformity of light collection efficiency and light yield on *irradiated* tiles

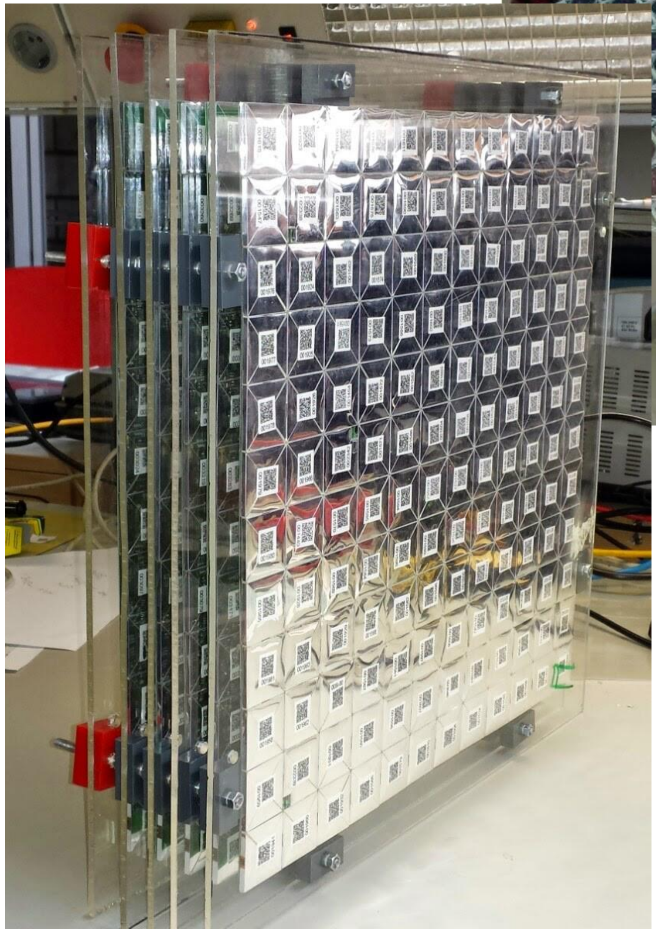
The CMS HGCAL

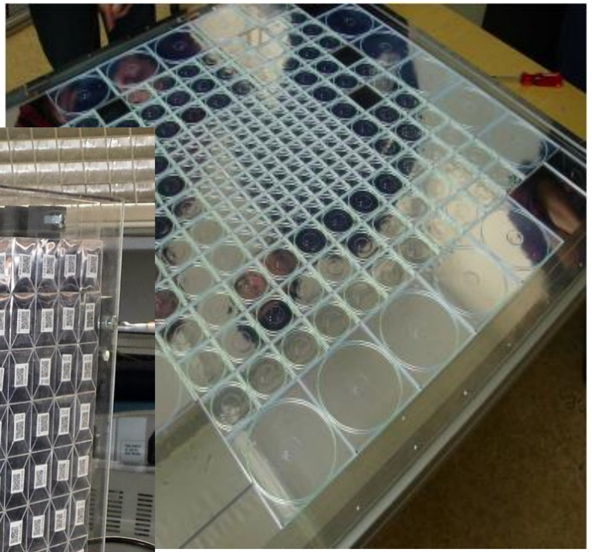
- LHC experiments will undergo an important upgrade in 2024-2026 (Phase-II Upgrades)
  - Necessary to overcome HL-LHC challenges: high interaction rate; significant radiation dose to be integrated over 10 years of operations
- CMS Phase-II Endcap calorimeter embraces the Particle-Flow approach to calorimetry
  - Design high-granularity detector to identify contribution from charged and neutral particles
- Key parameters of CMS HGCAL
  - 1.5<|η|<3.0
  - 600m^2 Si sensors; 500m^2 scintillator; 6M channels
- CE-E
  - Cu/CuW/Pb absorber; silicon sensors
  - 28 layers; 25X_0, ~1.3λ
- CE-H
  - Steel absorber silicon and scintillator
  - 24 layers; ~8.5λ



HGCAL Mechanical Layout





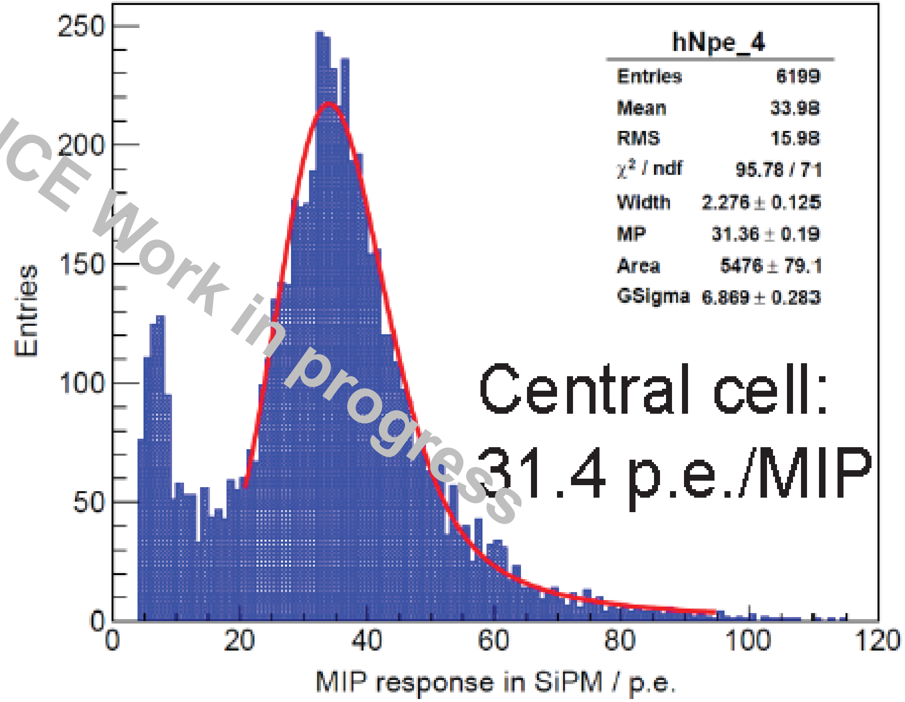


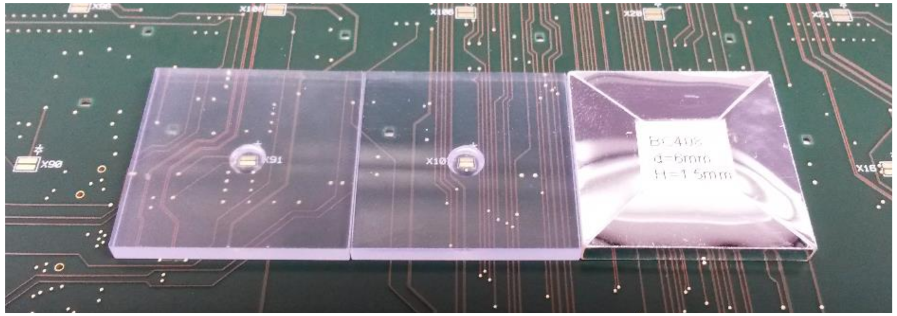
CALICE
AHCAL

Mixed Si-scintillator layer
Boundary optimized vs radiation hardness
Scintillator too in cold volume (-30C)

SiPM-on-Tile Setup

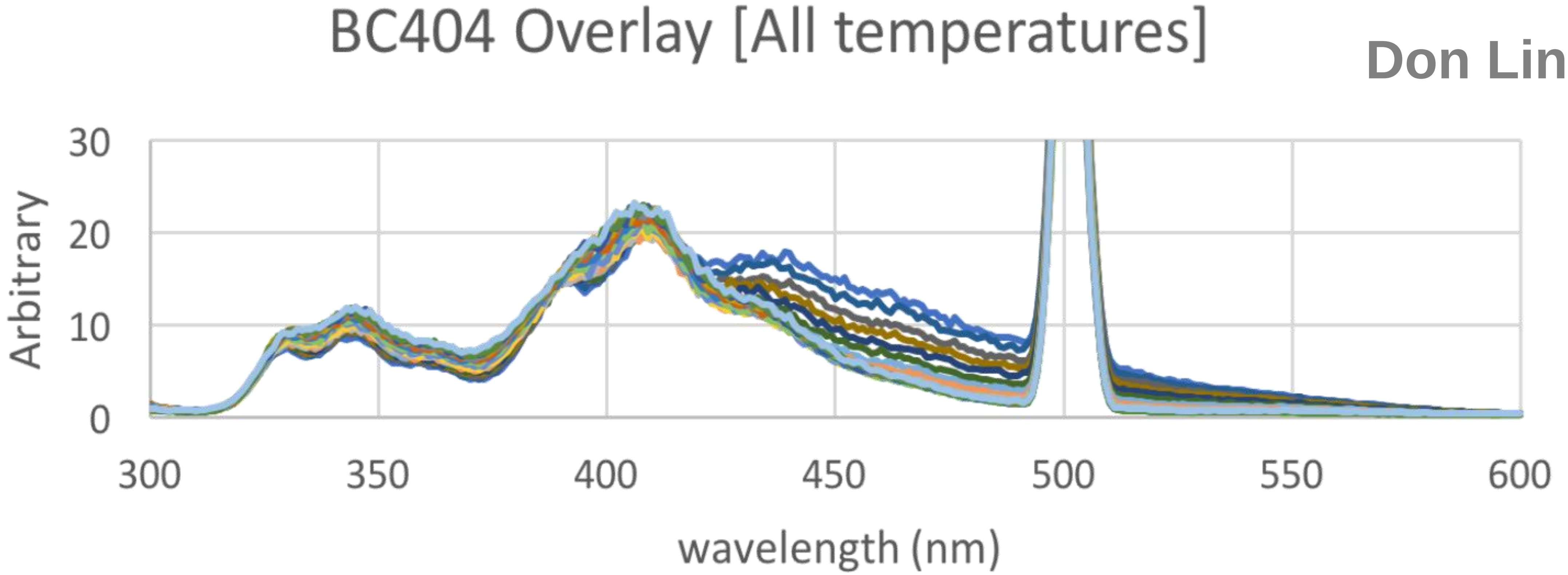
- Photosensor (SiPM) mounted directly on tile
  - Direct collection of scintillator light
  - Tile wrapped with reflective cover
  - Central dimple in tile optimizes light collection
- Cosmic-ray runs with prototype assembly
  - CALICE AHCAL prototype (similar structure to CE-H) tested at CERN Test Beam facility
- Scintillator tile and photosensor kept within the cold volume (-30C) in HGCAL design
  - Critical R&D question: how do scintillators behave when cold?





Cold Scintillators

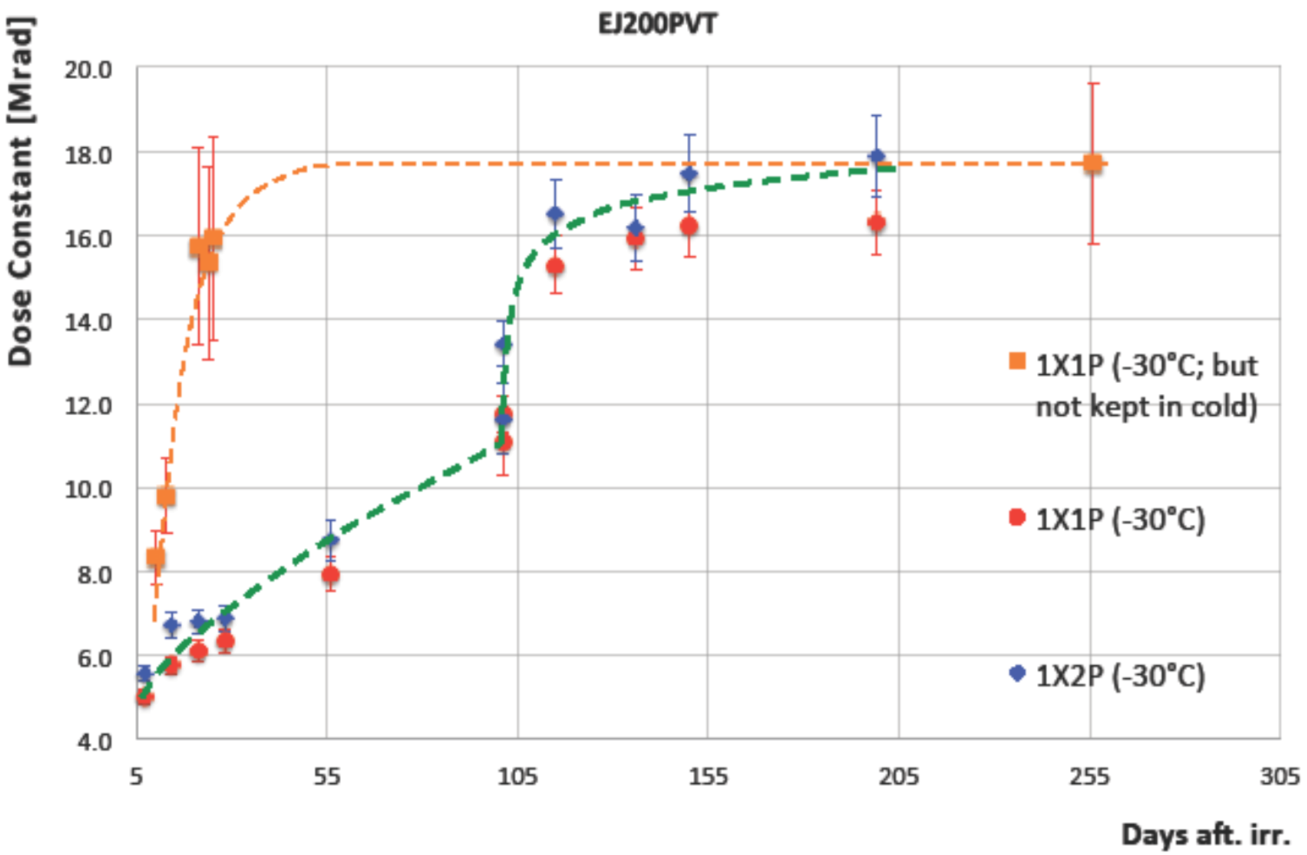
Don Lincoln



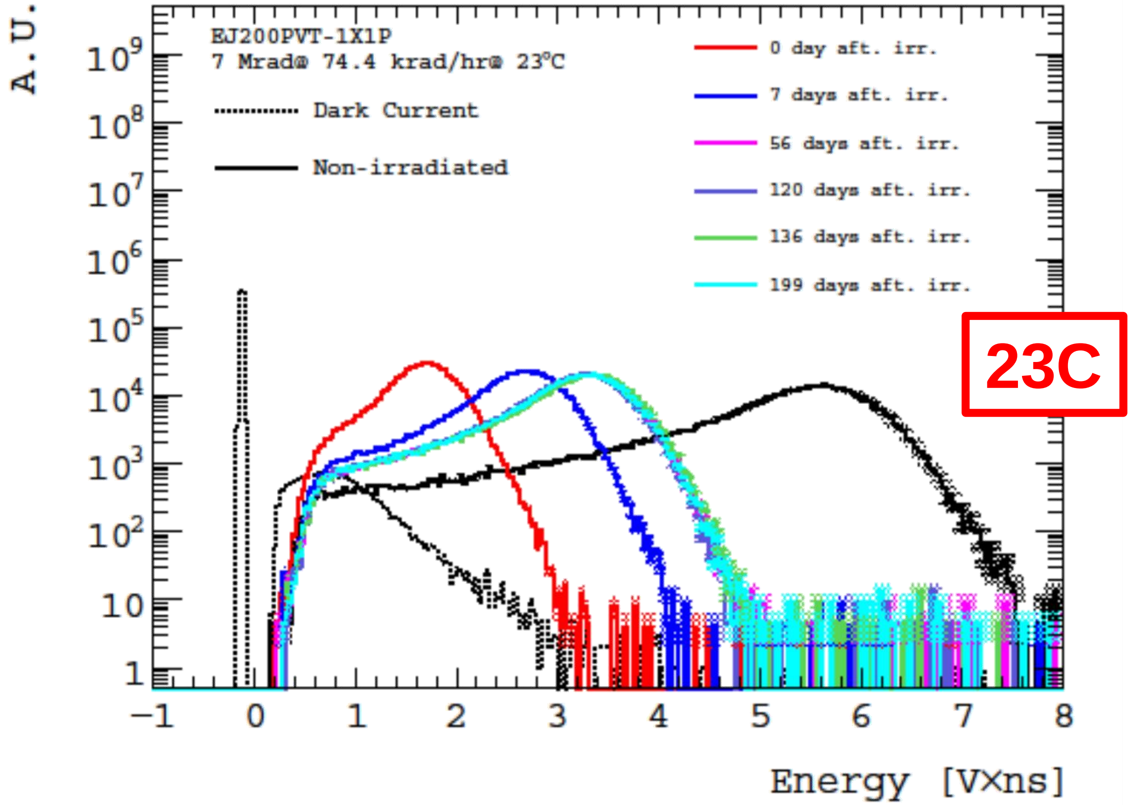
- Pulse shape and timing unaffected by temperature
  - Tested down to -180C (scintillator in liquid nitrogen)

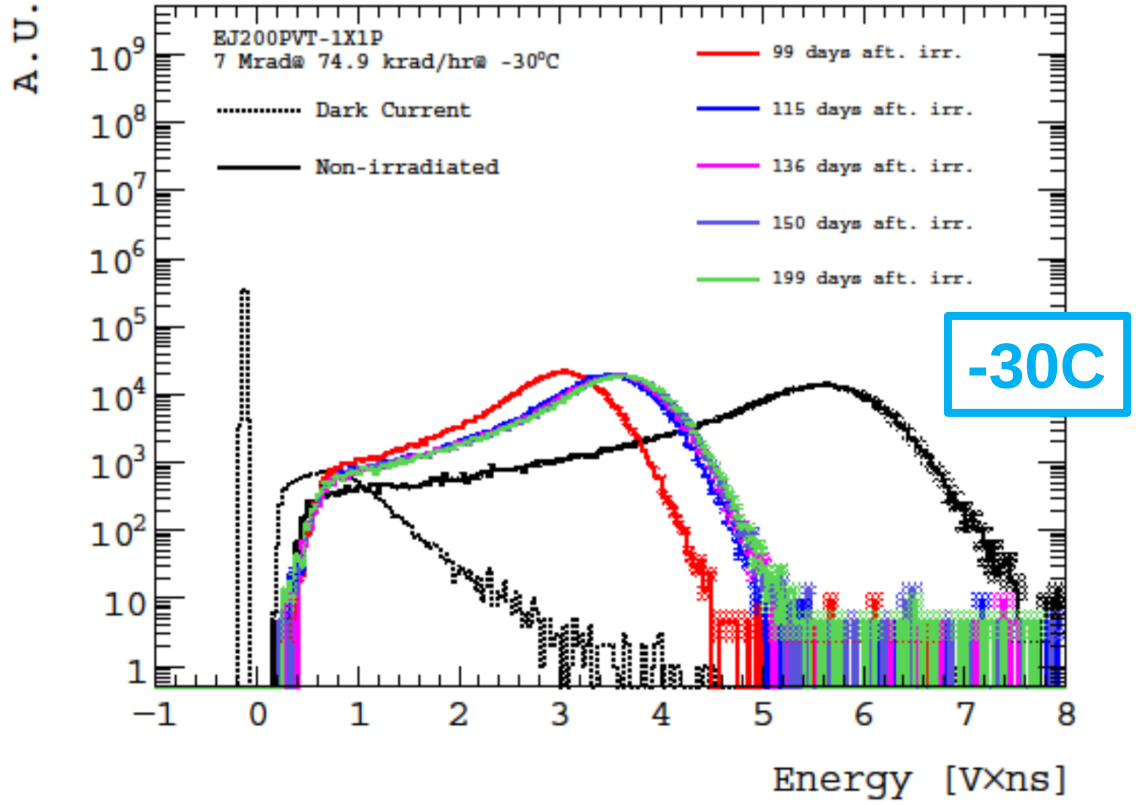
Cold Irradiation Annealing

- Monitoring annealing of damaged scintillator
  - 1x1x5cm^3 samples of plastic scintillator
  - Light yield with α-source
- Low temperature slows annealing, but no difference in permanent damage
  - Consistent with naïve expectation that creation of radicals and their reaction with diffused oxygen decrease with temperature



Cold vs. Warm Irradiation



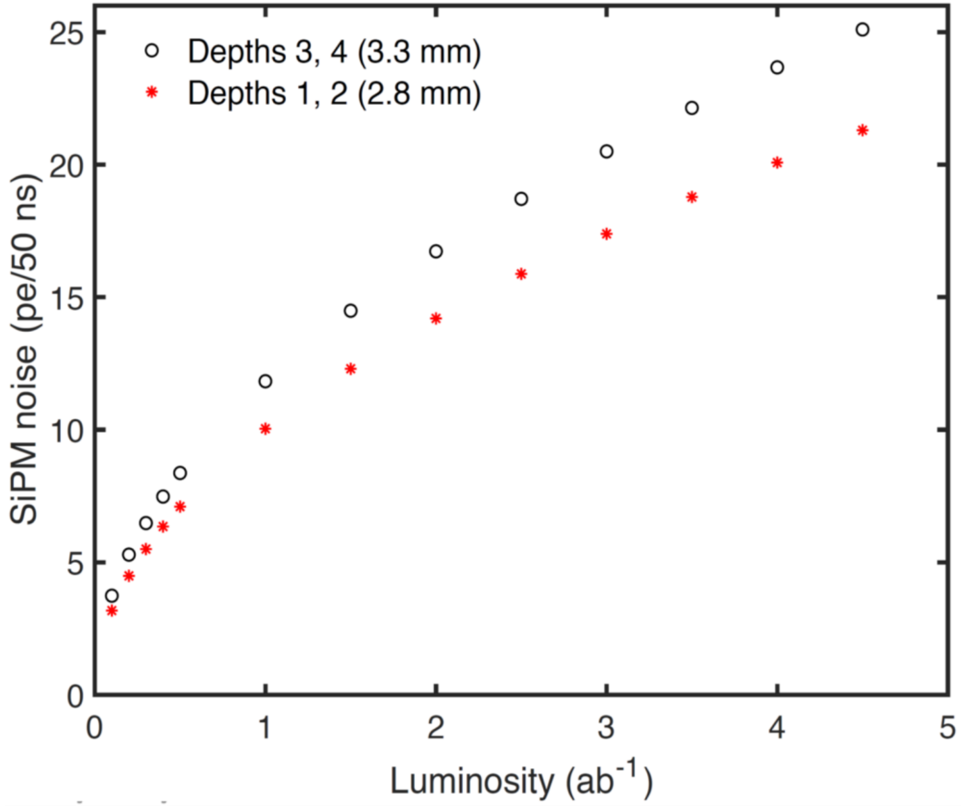


- Measured annealing (at room temperature) of samples irradiated at 23C and -30C
  - Indication that temporary damage anneals completely after ~4 months
  - Permanent damage is smaller in cold-irradiated samples

Photosensors Matter

Jim Hirschauer

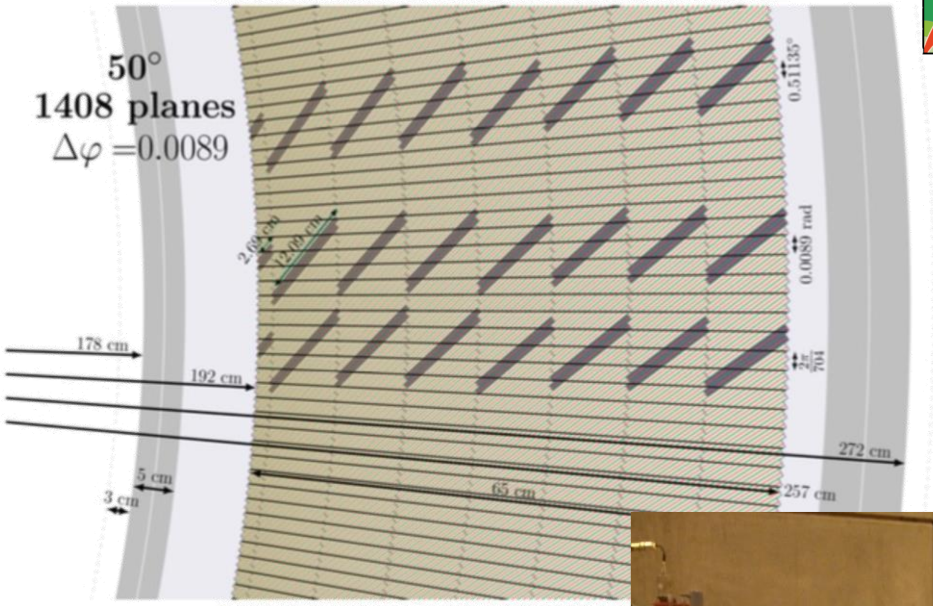
- Design of radiation-tolerant detector must include all components
  - – Lesson from CMS HE: observed light-yield reduction partially due to damage on photosensors (hybrid photo-diodes – HPD)
    - And another part to damage to wavelength-shifting fibers
- R&D effort devoted to characterizing radiation tolerance of photosensors
  - – Important contribution to detector design

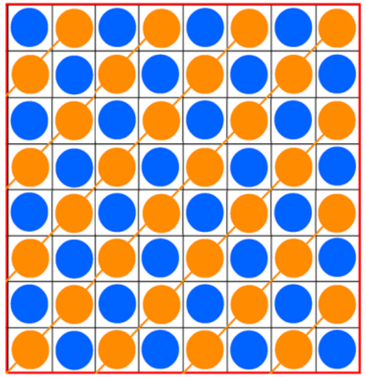


SiPM noise in 50ns gate
(~CMS Hadron Barrel light pulse)

A Look After HL-LHC

- Long-term prospective: 2030-2080
  - FCC-hh: 100km, 100TeV
  - ILC: 50km, e^+e^- at 1TeV
  - CEPC/SppC: 50km/100km, 100TeV
- Design of future detectors already started
  - R&D on granularity limits of noble liquid calorimeter
  - Dual-readout calorimetry





- Cherenkov
- Scintillator

Summary and Prospects

- Dose-rate effect and oxygen
  - Observe dependence based on dose rate, and diffusion depth
- Systematic study of radiation damage as a function of the scintillator composition
  - (Marginal) indications that emitting at longer wavelengths and increasing dopant concentration improve radiation tolerance
- Irradiations in cold environment (-30C)
  - Measurements do not seem to indicate cold is bad; on-going investigating at lower dose rates, to understand temperature dependence of oxygen diffusion, quenching of radicals, damage on dopants

- Modeling of radiation damage has multiple facets, with important correlations
  - Extent of damage, and type of damage, depends on integrate dose, dose rate, atmosphere (oxygen content and pressure), temperature, scintillator composition…
  - Literally years of measurements, converging toward set of publications

- Plastic scintillators are cheap, safe, and fit any detector design
  - Increasing their radiation tolerance can provide a good candidate material for large detectors where the expected integrated dose over the experiment lifetime is of the order of a few Mrad

Additional Material

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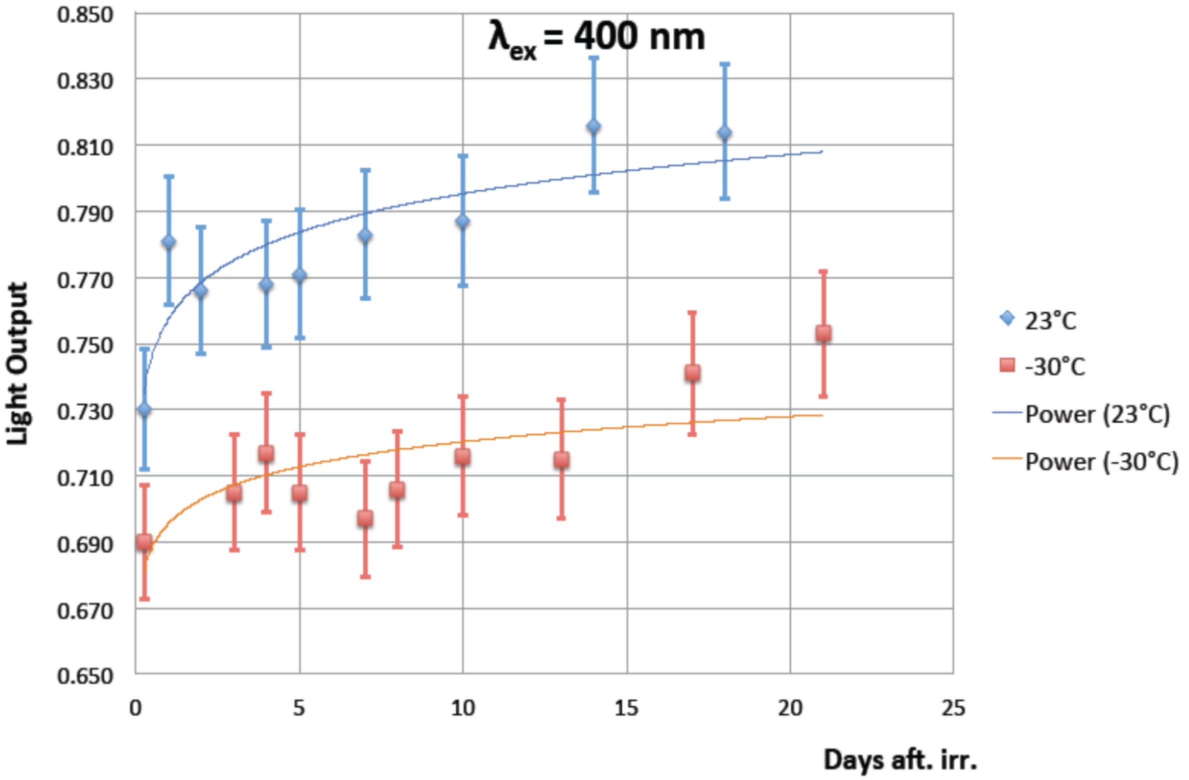
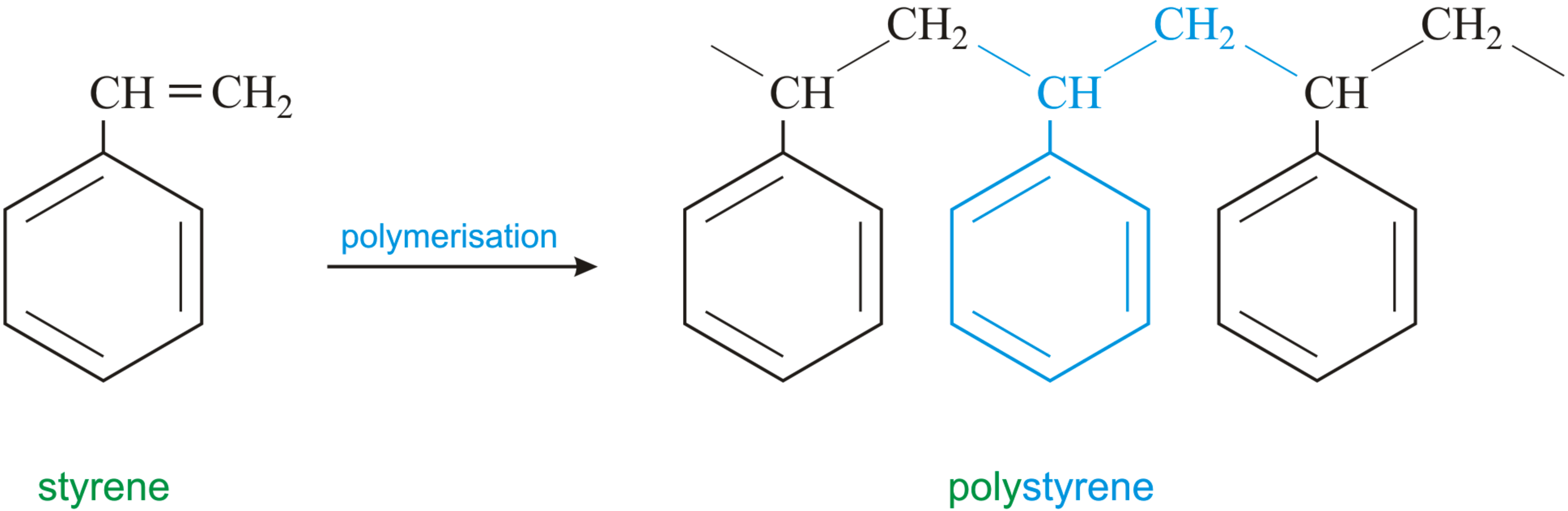
- Radiation induced oxidative degradation of polymers—I: Oxidation region in polymer films irradiated in oxygen under pressure; T. Seguchi, S. Hashimoto, K. Arakawa, N. Hayakawa, W. Kawakami, I. Kuriyama, Rad. Phys. Chem. 17 No 4, 1981

(Surface) Annealing

- Monitor evolution of ratio between integrals of emission spectra (irradiated vs. reference) to estimate annealing time
  - Emission measurement sensitive to (mostly) annealing of surface
  - Faster annealing time w.r.t. transmission measurements
    - Consistent with being sensitive exclusively to surface effect



Polymers



CH = CH2
polymerisation
CH2
CH2
CH2
CH
CH
CH
styrene
polystyrene